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X-ray studies of halloysite and metahalloysite.

Part I. The structure of metahalloysite, an example of a random layer lattice.

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METAHALLOYSITE is one of the kaolin group of minerals with a composition approximating to the formula $Al_2O_3.2SiO_2.2H_2O$, and its structure is generally considered to be built of composite layers of atoms of the kind found in other minerals of this group. It differs from them most strikingly in that it can be produced by the dehydration of the hydrated mineral halloysite, which has the approximate composition $Al_2O_3.2SiO_2.4H_2O$. Both minerals give comparatively poor X-ray diagrams showing relatively few lines and bands. The lines correspond to reflections from a basal spacing of about 7·2 Å. in metahalloysite and 10·1 Å. in halloysite. The similarity of the basal reflections from metahalloysite with those from the other kaolin minerals together with their similar chemical constitutions have provided the main evidence hitherto that metahalloysite is built from the same type of atomic layers. Hendricks (1942, p. 279) has stated that 'There is no reason at the present time for considering [meta] halloysite $Al_2O_3.2SiO_2.2H_2O$ to have types of layers different from kaolinite, nacrite, and dickite. New evidence, however, might be found that would lead to a more complete understanding of its structure. The powder-diffraction pattern of [meta] halloysite differs somewhat from that of kaolinite, but in a manner that can be accounted for by its degree of organization', and later he states that there is great lack of order in the stacking of the layers in both metahalloysite and halloysite. It is this question of the disorderliness

in the stacking of the layers which will be mainly considered in the present paper.

The present writers with MacEwan (1946) have already suggested in a brief note that the layers may be assembled in so random a manner in their own planes that the diffraction effects which are observed may be produced by two-dimensional sheet-like gratings, apart from the basal reflections which arise from the stacking of the layers. The evidence for this view was that the form of the most prominent band in the diffraction pattern could be explained by a simple application of the theory developed by Warren (1941) for X-ray diffraction by two-dimensional lattices. The bands, however, are of various forms and require more detailed consideration than was possible in the previous note.

The diffraction bands (not the basal reflections) from halloysite and metahalloysite appear to be identical; no change can be observed when halloysite is dehydrated to metahalloysite. This indeed would be expected if in both cases the bands arose from the same randomly displaced layers. We have preferred to examine in detail the phenomena obtained with metahalloysite partly because the basal reflections can be sharpened by heat-treatment, which, however, does not affect the diffraction bands, and also because the smaller basal spacing gives fewer reflections of this type.

Comparatively little attention has been given to the structure of metahalloysite, probably because the diffraction pattern is poor and provides unpromising material for detailed study. Nevertheless the question is worth examining in as much detail as possible in order to obtain the clearest possible picture of the difference between kaolinite and metahalloysite and their relation to other kaolin-type minerals, such as those which commonly occur in many fireclays (*see* Brindley and Robinson 1946b, 1947) and which appear to have intermediate degrees of orderliness.

The X-ray powder diagram of metahalloysite.

X-ray diagrams of a number of metahalloysites have been taken with Cu-$K\alpha$ radiation in a camera of 20 cm. diameter of the semi-focusing type, employing a flat plate of powder; a few measurements have also been made with a camera of 12 cm. diameter using a fine rod of powder. The measurements were made on metahalloysite from Lawrence County, Missouri, U.S.A., which was the purest natural material available when

this work was begun. Subsequently, however, we suspected that two lines in this diagram were spurious and further data, more particularly the photometric measurements, have been based on a natural metahalloysite from Simla, India, which does not give these two suspected lines. A number of artificial metahalloysites prepared by dehydration

TABLE I. Lattice spacings (in Å.) for specimens of metahalloysite.

I.	II.	III.	IV.	V.
7·69	7·40	7·55	7·48	7·46
4·422	4·423	4·42	4·44	4·45 4·41
3·578	3·596	3·66	3·62	3·63
2·559	2·557	2·56	2·60	2·55
—	2·486+	—	—	—
2·403	2·400	—	—	—
—	2·335+	2·36	2·33	2·32
2·218	trace	2·23	—	—
—	—	2·06	—	—
—	1·801*	—	—	—
1·678	1·677	1·678	1·70 1·64	1·67
1·481	1·481	1·481	1·487	1·48
1·281	1·281	1·283	1·290	1·27
1·231	1·231	1·232	1·237	1·23
—	1·201*	—	—	—
—	1·108	1·105	—	—
—	1·021	1·021	—	—
—	0·968	0·968 0·958	—	—
—	0·856	0·855	—	—
—	0·840	0·838	—	—

* Observed only after heating for several hours at 300° C.
\+ Doubtful metahalloysite lines.

I. Specimen from Simla, India (B.M. 43087).
II. Specimens from Lawrence County, Missouri, U.S.A.
III. Data from D. M. C. MacEwan (specimen from Hungary).
IV. Data by G. Nagelschmidt (1934) (specimen from Harz, Germany).
V. Data by M. Mehmel (1935).

of halloysites from Angleur (Belgium), Northamptonshire (England), and Indiana and Utah (U.S.A.), have also been examined and these give results in agreement with the Simla material; the last two, however, contain gibbsite as an impurity. In table I we record the data for the Missouri and Simla specimens; the higher orders, which owing to their diffuseness are difficult to measure accurately, have been determined only for the Missouri material and the data in the table are mean values from a number of films. The table also contains data supplied privately

by Dr. MacEwan for a Hungarian halloysite together with data by Nagelschmidt (1934) and by Mehmel (1935). There is good general agreement between the results, and especially between the higher order spacings obtained by MacEwan and ourselves. The two doubtful lines in the Missouri pattern, marked + in table I, occur in a range of continuous scattering for which most previous workers have recorded only the approximate beginning and end of the band. We find for all metahalloysites evidence of two intensity maxima in this band.

Indexing of the powder diagram.

The diffraction pattern consists of lines and bands which may conveniently be considered separately. A microphotometer trace of the

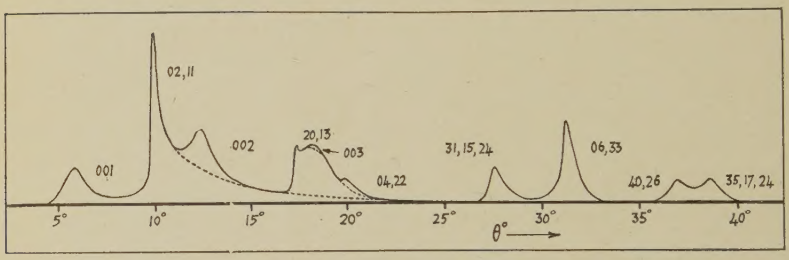

FIG. 1. Microphotometer trace of X-ray powder diagram of metahalloysite; X-ray intensity plotted against Bragg angle θ.

lower-order reflections (fig. 1) shows an almost continuous distribution of intensity across the film and the lines are distinguished from the bands only by having a more symmetrical distribution of intensity.

(*a*) *The basal reflections*, (00*l*).—These consist of lines which can be indexed as various orders of (001) with a spacing of about 7·2 Å. They are listed in table II together with the corresponding kaolinite reflections (Brindley and Robinson, 1946a). The greater breadth and more rapid diminution of intensity towards higher angles of the metahalloysite reflections indicates smaller crystals and/or less perfect parallelism of the layers; probably both causes are operative. In part III of this group of papers the effect of heat-treatment on metahalloysite is considered; heat-treatment sharpens the basal reflections and makes it possible to observe some of the higher orders which normally are not visible. Moreover, the observed spacings then agree better with submultiples of the fundamental spacing of 7·20 Å. There appears to be no doubt that the basal spacing of metahalloysite is somewhat greater than that of kaolinite.

TABLE II. Comparison of basal reflections (00*l*) from metahalloysite and kaolinite. d_{001} (metahalloysite) = 7·20 Å., d_{001} (kaolinite) = 7·132 Å.

	Metahalloysite (Missouri).			Kaolinite.		
00*l*.	*d* (obs.).	*d* (calc.).	*I* (obs.).	*d* (obs.).	*d* (calc.).	*I* (obs.).
001	7·40	7·200	vst	7·15	7·132	vvst
002	3·596	3·600	vst	3·566	3·566	vvst
003	2·400	2·400	w	2·374	2·377	mst
004	1·801*	1·800	w	1·778	1·783	m
005	—	1·440	abs.	1·426	1·426	w-m
006	1·201*	1·200	vw	1·188	1·189	w-m
007	—	1·029	abs.	1·019	1·019	w

* Observed only after heating for several hours at 300° C. Visual intensity scale: vvst, vst, st, mst, m, w-m, w, vw, abs.

(*b*) *Diffraction bands, with indices* (*hk*).—These bands are of considerable interest for their intensity distribution varies noticeably from one to another. The lowest-order band is the most prominent feature of the whole powder diagram; it has a sharp low-angle termination with a maximum of intensity corresponding to a spacing of 4·42 Å. while on the high-angle side the diffracted intensity spreads well beyond the (002) reflection with $d = 3{\cdot}60$ Å. The present writers with MacEwan (1946) have already shown that the variation of intensity in this band agrees well with the theories of diffraction by two-dimensional crystal lattices developed by Laue (1932) and Warren (1941). This preliminary study assumed the lattice-structure factor was constant over the range of angles covered by a band; in the present paper it will be shown that, while the assumption was valid for this particular band, it is not generally valid. A more searching test of the supposition that the bands are produced by two-dimensional diffraction requires that both the forms and the relative intensities of all the bands in the powder diagram should be explained on this basis.

Consideration will first be given to the *positions* of the bands and their indices. The intensity maxima would be expected to occur approximately in directions θ_0 corresponding to Bragg reflections from 'planes' (*hk*) having a spacing given by

$$d_{hk} = [(h/a)^2+(k/b)^2]^{-\frac{1}{2}}, \qquad (1)$$

where a and b are the lattice parameters of the layer structure. The extension of Laue's theory by Warren (1941) takes account of the finite size of the sheets. When the linear extension L of the crystallites is small, e.g. of the order of 100 Å., the intensity maxima are displaced from θ_0 towards higher angles and the apparent spacings d' derived from

the observed maxima are less than the true spacings d_{hk} by a correction term Δd given approximately by

$$\Delta d = d_{hk} - d' = 0{\cdot}32 d_{hk}^2/L. \qquad (2)$$

Such displacements have been observed by Biscoe and Warren (1942) and by A. Taylor (1942) for carbon black, and by Aruja (1944) for chrysotile, a mineral structurally similar to the kaolin group but containing magnesium in the octahedral positions.

TABLE III. Two-dimensional X-ray diffraction maxima from metahalloysite (Missouri).

d_{hk} calculated with $a = 5{\cdot}14$ Å., $b = 8{\cdot}90$ Å., $b/a = \sqrt{3}$.
Δd calculated from equation (2) with $L = 200$ Å.

(hk).	d' observed.	Δd.	$d_{hk} = d' + \Delta d$.	d_{hk} calc.	Intensity observed.
11,02	4·423	0·031	4·454	4·451	vvs
20,13	2·557	0·011	2·568	2·570	s
04,22	(2·218)*	0·008	2·226	2·225	vw
24,31,15	1·677	0·004	1·681	1·682	s
33,06	1·481	0·003	1·484	1·484	vs
40,26	1·281	0·002	1·283	1·285	ms
42,35,17	1·231	0·002	1·233	1·234	ms
08,44	1·108	0·002	1·110	1·112	w
51,28,37	1·021	0·002	1·023	1·021	vw
53,46,19	0·968	0·002	0·970	0·971	vw
55,0 10	**	—	—	0·890	—
60,39	0·856	0·001	0·857	0·857	w
62,2 10,48	0·840	0·001	0·841	0·841	vw

* Observed, but not accurately measured from Missouri halloysite. The value 2·218 obtained with a specimen from Simla, India.

** Not observed.

Visual intensity scale: vvs, vs, s, ms, m, wm, w, vw.

As table III shows, we also find evidence for measurable displacements of the intensity maxima of metahalloysite from the positions calculated by means of equation (1). In this table, d' is the directly observed spacing, Δd the correction term calculated for a crystallite size $L = 200$ Å. (see later for the determination of L), and $(d' + \Delta d)$ is the 'experimental' value of d_{hk}. The latter may be compared with d_{hk} calculated by means of equation (1) with the following lattice constants:

$$a = 5{\cdot}14 \text{ Å.}, \quad b = 8{\cdot}90 \text{ Å.}$$

These parameters are practically the same as for kaolinite, viz., $a = 5{\cdot}14$ Å., $b = 8{\cdot}93$ Å., but whereas in kaolinite a is unquestionably less than $b/\sqrt{3} = 5{\cdot}155$ Å., in metahalloysite the reflections are not sufficiently well defined or numerous to decide whether this is the case or not. We have therefore taken $a = b/\sqrt{3}$. The table shows close

agreement between the experimental and calculated values of d_{hk} only when the correction term Δd is incorporated.

(*c*) *The full powder diagram.*—All the observed reflections from metahalloysite can be indexed as basal (00*l*) reflections or as (*hk*) bands. In particular there are no reflections of type (*hkl*). This result is fully in accord with the general view of the metahalloysite structure described in the opening paragraphs. Absence of (*hkl*) reflections implies complete lack of order in the stacking of the layers, or such a high degree of disorder that (*hkl*) reflections are of negligible intensity. The two doubtful reflections from the Missouri metahalloysite at spacings of 2·486 and 2·355 Å. may be briefly considered; they agree closely with two prominent lines in the kaolinite diagram, viz., a strong reflection at 2·486 Å. and a very strong reflection at 2·331 Å. No trace, however, is found of an equally strong kaolinite line at 2·284 Å., or of other kaolinite lines which might have been expected if the two doubtful lines arose either from kaolinite impurity or from a partial ordering of the layers which was producing effectively regions of kaolinite within a metahalloysite matrix. If partial orderliness should occur, then the type found in many fireclays (cf. Brindley and Robinson, 1947) corresponding to random displacements of layers parallel to the *b*-axis only might have been expected. There is no evidence, however, for such ordering of the layers, for the fireclay type of kaolin mineral gives a medium-strong line at 2·50 Å. rather than a line at 2·486 Å., while there is a strong and rather broad line at 2·325 Å. as compared with 2·331 Å. in kaolinite and 2·335 Å. for the doubtful line in the Missouri metahalloysite. The origin of the two extra lines in the Missouri metahalloysite pattern must therefore be left open for the present; they are unlikely to arise from kaolinite impurity or from a partial ordering of the 'fireclay type' and there is insufficient evidence that they arise from an ordering of the kaolinite type. This uncertainty, however, is concerned only with the Missouri material, and the other metahalloysites we have examined fully support the statement that the only reflections are of types (00*l*) and (*hk*).

Distribution of intensity in the (hk) bands.

(*a*) *Theoretical calculation.*—Warren's treatment of the problem of diffraction by two-dimensional lattices involves certain simplifying assumptions which appear to be justified, at least to a first approximation.[1] He shows that the intensity $I_{2\theta}$ diffracted in an (*hk*)

[1] Further work by A. J. C. Wilson and by G. W. Bradley and J. Méring (Nature, London, 1948, vol. 161, pp. 773–775 [M.A. 10–363]) tends to confirm the approximations used by Warren, but certain features, such as the structure of the 20,13 band in fig. 3, may not be given correctly by the approximate treatment.

band in direction 2θ with respect to the incident beam is given by

$$I_{2\theta} = C\Phi(\theta) \sum F^2, \tag{3}$$

where C is a constant, F is the structure factor for the unit cell of the layer and the summation extends over all reflections contributing to a band, and $\Phi(\theta)$ is given by the following expression:

$$\Phi(\theta) = 2\left(\frac{L}{\sqrt{\pi}\lambda}\right)^{\frac{1}{2}} \frac{1+\cos^2 2\theta}{(\sin\theta)^{\frac{3}{2}}} f(a), \tag{4}$$

where $f(a)$ is an integral evaluated numerically by Warren and

$$a = 2\sqrt{\pi}.\ L(\sin\theta - \sin\theta_0)/\lambda.$$

For $a > 3$, the following approximation is useful:

$$\Phi(\theta)_{a>3} \doteq \frac{1+\cos^2 2\theta}{\sin\theta(\sin^2\theta - \sin^2\theta_0)^{\frac{1}{2}}}.$$

The variation of F with angle θ is most conveniently calculated in terms of an artificial third index l' given by

$$l' = \frac{2c}{\lambda}(\sin^2\theta - \sin^2\theta_0)^{\frac{1}{2}}, \tag{5}$$

where c can be taken as the basal spacing, 7·20 Å. It must be emphasized that this amounts to no more than a convenient method of calculating the continuous variation of F with θ and it does not imply the usual three-dimensional diffraction process; in this equation l' can have non-integral values corresponding to any chosen values of $\sin\theta$.

The calculation of F as a function of θ for each diffracted band requires a knowledge of the structure of the layer. This has been taken to be the same as in kaolinite (Brindley and Robinson, 1946a). Although the cell has ortho-hexagonal shape, the atomic distribution does not possess hexagonal symmetry. In consequence, F^2 must be calculated for each index pair (hk) contributing to a band and this is indicated in equation (3) by the summation sign.

In order to evaluate I as a function of θ by means of equations (3), (4), and (5), the linear dimension L, the extension of the crystal layer, must be known. L may in fact be different for different reflections depending on the habit of growth of the crystalline sheets. There are various ways of obtaining an approximate value of L, but ultimately the best procedure in order to avoid unnecessary and rather doubtful approximations is to calculate I as a function of θ for a range of L values and then make comparison with the observed diffraction bands. This

procedure is long and tedious, but since the Laue–Warren theory has not yet been tested by detailed comparison with experimental results, it seems worth while to make such a comparison in at least one case.

Warren shows that if F can be treated as effectively constant in equation (3) so that I depends only on $\Phi(\theta)$, then L is related to the half-breadth B of the band by the approximate relation

$$L = 1{\cdot}84\lambda/B\cos\theta, \qquad (6)$$

an equation closely resembling Scherrer's equation for three-dimensional crystals. When this equation is applied to the observed half-breadths of the diffraction bands from metahalloysite after systematic correction for $K\alpha_1\alpha_2$ separation and for the focusing properties of the X-ray camera, values of L are obtained ranging from about 100–250 Å. This range of values arises, at least in part, from the quite considerable variation of F with θ for some of the bands, and it is only for those bands with approximately constant F values that L can be reliably obtained. L can also be estimated by applying equation (2); if the lattice constants a and b are determined from high-order reflections for which Δd in any case is small, then the difference between d_{hk} calculated for low-order reflections and d' observed experimentally enables L to be found. The method is again approximate and is also inaccurate because $(d_{hk}-d')$ is always fairly small.

Despite the inaccuracies and approximations of these methods, they suffice to give the order of magnitude of L, which is found to be about 100–300 Å. We have therefore calculated fully by means of equations (3), (4), and (5) the variation of I with θ for the six most prominent bands in the diffraction pattern, taking $L = 100$, 200, and 300 Å.

(*b*) *Comparison of observed and calculated intensity curves.*—The results of the calculations and their comparison with experimental data are shown in figs. 2 and 3. In the first place fig. 2*a* shows the variation of the angular function $\Phi(\theta)$ with θ for the 02,11 reflection and $L = 200$ Å.; this curve corresponds to the intensity distribution, I plotted against θ, which results when ΣF^2 is constant or nearly constant over the angular range of a band. Figs. 2*b*–2*g* show ΣF^2 plotted against θ and against the artificial third index l' of equation (5). In Warren's treatment of the problem, the calculated I,θ curves are obtained for each band and for each value of L by multiplying the appropriate $\Phi(\theta)$ curve of the type shown in fig. 2*a* by the ΣF^2 curve. The resulting intensity curves, scaled to the same peak intensity for each reflection, are shown in fig. 3*a*–3*e*.

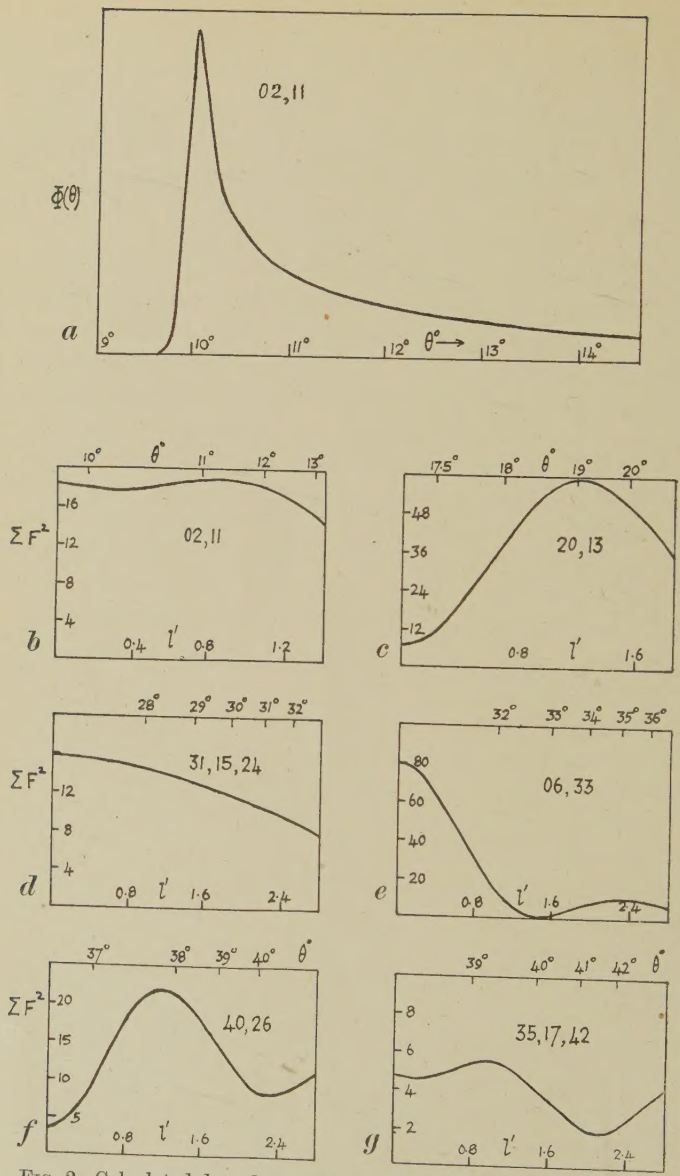

FIG. 2. Calculated data for two-dimensional diffraction bands from metahalloysite.

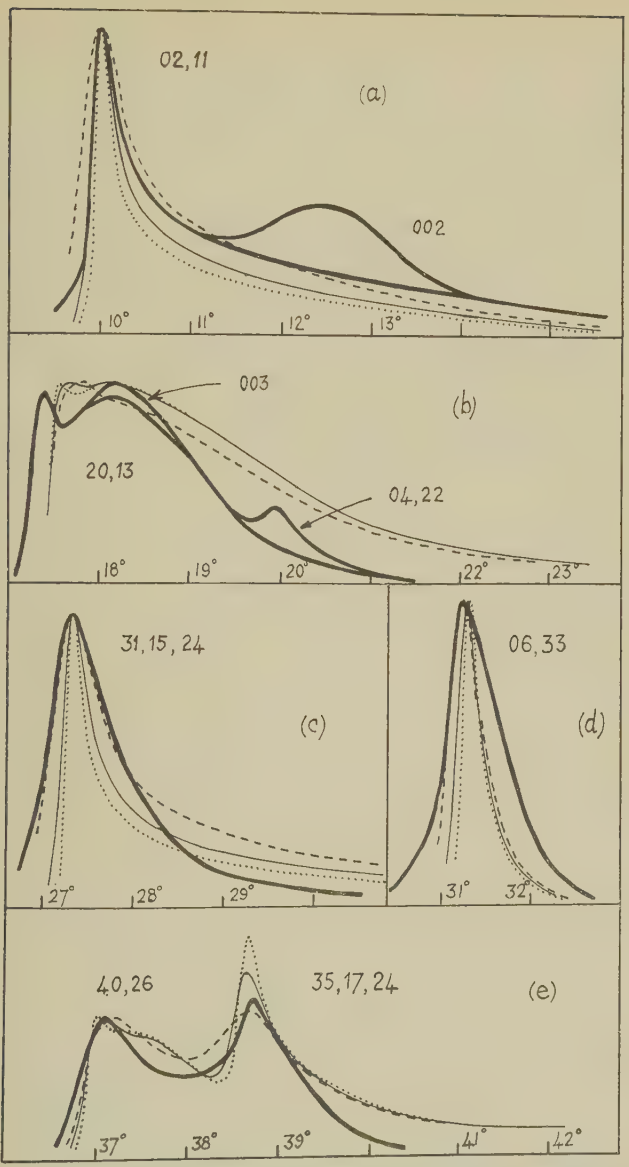

FIG. 3. Comparison of observed and calculated diffraction bands from metahalloysite. Heavy curves show experimental results. Light curves show calculated results for different crystal sizes, L; --- for $L = 100$ Å.; —— for $L = 200$ Å.; .... for $L = 300$ Å.

The experimental curves obtained by careful microphotometry of the X-ray diagrams are shown in fig. 3*a*–3*e* by heavy curves. Where reflections are superimposed, as in figs. 3*a* and 3*b*, we have endeavoured to reconstruct the separate peaks, but in the case of the overlapping bands (40,26) and (35,17,24) we have preferred to treat the two as a composite group.

It is seen at a glance that the calculations reproduce the main features of the observed curves and furthermore it is evident that the variety of bands which are observed arises from the variety of ΣF^2 curves. Thus the (02,11) band and the (31,15,24) band closely resemble the $\Phi(\theta)$ curve of fig. 2*a* because in both cases ΣF^2 is largely constant. On the other hand, the third-order band (06,33) is comparatively sharp because ΣF^2 shows a very rapid diminution as θ increases. Conversely the broad, almost flat-topped (20,13) band is produced by a rapidly rising ΣF^2 curve. As regards the combined (40,26), (35,17,24) band, the second peak is the sharper as a consequence of the approximate constancy of ΣF^2 for this peak, while the rapid increase of ΣF^2 for the (40,26) band tends to produce a broad band of the (20,13) type.

While the Laue–Warren theory clearly goes a long way towards explaining the observed results, we have not succeeded in explaining the finer details of the observed results. There is, for example, no single value of L which will account quantitatively for the observed widths of the bands. Some differences between L values for different bands might be expected owing to the shape of the crystalline units, but no satisfactory explanation of the observed results has been obtained along these lines. In particular we see that the (02,11) and (06,33) bands are not consistent with a single L value. The observed (06,33) band appears to be broad compared with the (02,11), a result which may arise from some separation of the components bands, (06) and (33), which would occur if the b parameter were not exactly $\sqrt{3}.a$. The experimental data, however, scarcely justify detailed discussion of this point.

The observed peaks in fig. 3 owe their widths partly to separation of the $K\alpha_1\alpha_2$ components—a small effect—and to the focusing properties of the X-ray camera. Both these questions have been examined in detail. They do not affect materially the general conclusions drawn above, but when allowance is systematically made for them, we find that the observed widths of the bands indicate a crystal size L of the order of 150–200 Å.

(*c*) *Comparison of observed and calculated peak intensities.*—Owing to the broadness of the bands, it is more convenient to compare peak

intensities than integrated intensities. The calculated values depend on the crystal size L and the detailed numerical work shows that the dependence of peak height on L varies with the type of band. The main interest in considering the relative peak intensities is the evidence it may provide for or against the kaolinite type of structure for the layers in metahalloysite. Further evidence regarding L may also be obtained. Table IV compares the observed and calculated peak intensities expressed relative to 100 for the 20,13 band, for L = 100, 200, and 300 Å. The effect of absorption in the powder specimen has been taken fully into account.

TABLE IV. Comparison of observed and calculated peak intensities in diffraction bands.

	Calculated peak intensities for			Observed peak
(*hk*).	L = 100 Å.	L = 200 Å.	L = 300 Å.	intensities.
02,11	147	228	274	337
20,13	100	100	100	100
04,22	15	24	32	20
31,15,24	51	77	95	72
06,33	140	223	274	157
40,26	22	29	22	46
35,17,24	22	39	35	47

Consideration of the data in the table shows that an L value of the order of 150–200 Å. gives general agreement between the observed and calculated values. This not only confirms the previous value of L, but shows that the layer structure is almost certainly the same as in kaolinite.

Summary and conclusions.

The paper provides a more extensive range of powder data for metahalloysite than has previously been published. The reflections are divided into basal reflections of type (00*l*) and bands of type (*hk*). The latter arise from X-ray diffraction by the two-dimensional regularity within the kaolin-type layers. No reflections of type (*hkl*) are observed. The lattice dimensions are $b = 8{\cdot}90$ and $a = b/\sqrt{3} = 5{\cdot}14$ Å. Possible evidence is considered that a may not be exactly equal to $b/\sqrt{3}$; while a and b are practically the same as for kaolinite, the layer spacing is 7·20 Å. which is slightly greater than 7·132 Å. for kaolinite.

Quantitative data based on the Laue–Warren theory of diffraction by two-dimensional lattices are given supporting the view that there are random displacements between successive kaolin layers. The band widths and the peak intensities are generally accounted for by assuming

(i) a layer structure of the type found in kaolinite, and (ii) a crystal size for the layer units of 150–200 Å. in the plane of the layer. The importance of taking account of the variation of structure factor F with angle θ within the range covered by a single band is specially emphasized and the variety of band types exhibited by metahalloysite is shown to arise directly from the manner in which ΣF^2 varies with θ (cf. figs. 2 and 3).

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X-ray studies of halloysite and metahalloysite.

Part II. The transition of halloysite to metahalloysite in relation to relative humidity.

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THE experiments described below on the dehydration of halloysite were largely carried out in the Laboratoire Central des Services Chimiques de l'État, Paris, where we enjoyed not only the excellent facilities of the laboratory but also the advice of Monsieur J. Méring and Mlle R. Glaser who had made similar studies of montmorillonite. The use of a Guinier-type focusing camera with strictly monochromatic radiation was especially useful. These experiments were undertaken because little was known about the dehydration process beyond the recognition that it occurs very readily in dry atmospheres and at low temperatures. The main experimental difficulty lies in differentiating between water adsorbed on external surfaces of the clay particles (adsorbed water) and water internally absorbed between the kaolin layers (interlayer water). Experiments by Alexander *et al.* (1943) in which the changes of weight were determined when halloysite was brought to equilibrium firstly at 75% humidity, then at a lower humidity, and finally at 75% humidity again were difficult to interpret, as they themselves recognized. They also dried the mineral at 300° C. before returning it to 75% humidity and found that only about half the total water lost was regained, from which it was clear that the adsorbed and interlayer water contents were of comparable importance.

Our own experiments have followed up this latter line of inquiry and parallel X-ray studies have shed considerable light on the mechanism of the processes which occur. The halloysite used also came from Eureka, Utah, which was the source of the material used by Alexander *et al.* Weighing experiments were carried out as follows: Two equal samples of halloysite of about 0·5 gram weight were prepared, one of which was

then heated to 300° C. for 2–3 hours to remove adsorbed and interlayer water, cooled in a desiccator and reweighed. Let w_1 be the initial mass, w_2 the mass after heating and cooling. The two samples were now placed over a solution of H_2SO_4 and H_2O adjusted to give the required relative humidity. After two days and again after four days they were reweighed; generally there was no significant difference between these weights, so that presumably equilibrium had been attained. If w_3 is the new mass of the unheated sample and w_4 that of the heated sample, then (w_3-w_4) will measure the interlayer water corresponding to the chosen humidity, provided (*a*) no interlayer water returns to the lattice after baking at 300° C., and (*b*) the adsorbed water is the same for the heated and unheated samples. X-ray evidence indicates that (*a*) is true or very nearly true.

If any interlayer water should return to the heated material, then (w_3-w_4) will be *less* than the mass of interlayer water at the chosen humidity; we return to this point later when discussing results. As regards (*b*), provided the heat-treatment does not change the total surface area, the adsorbed water at any chosen humidity will probably not be changed. With this assumption, (w_4-w_2) measures the adsorbed water.

An additional complication arises in practice because the original material contains a small percentage of gibbsite, $Al_2O_3.3H_2O$, which dehydrates to boehmite, $Al_2O_3.H_2O$, at about 200° C. Alexander *et al.* make no reference to gibbsite in their Utah halloysite though they mention it as a possible impurity in halloysite used by Hofmann, Endell, and Wilm (1934). We have estimated the gibbsite impurity from X-ray intensity measurements to be $3{\cdot}6\pm0{\cdot}7$ %, and this has been confirmed by Dr. R. W. Grimshaw who by thermal analysis methods has obtained $2{\cdot}9\pm0{\cdot}2$ %. The original material also contains an admixture of metahalloysite, the effect of which on the interpretation of the results will be considered later.

Results of the dehydration measurements.

These are set out in table I and are scaled to correspond to an initial sample of exactly 0·500 gram. They have been corrected systematically for the presence of 3·6 % gibbsite; the original mass, w_1, therefore appears as $0{\cdot}500-0{\cdot}018 = 0{\cdot}482$ gram. The table itself is self-explanatory.

The following features are shown more clearly in fig. 1: as the humidity increases the adsorbed water (curve *a*) rises from almost zero rapidly at first but more slowly at higher humidities. The interlayer water (curve *b*) rises slowly at first from a value *not equal to zero* and more

TABLE I. Dehydration data for halloysite (Eureka, Utah).

w_1 = mass of original sample = $0{\cdot}482_0$ gram.
w_2 = mass of sample after baking at 300° C. = $0{\cdot}419_7$ gram.
w_3 = mass of unheated sample after standing in relative humidity, R %.
w_4 = mass of heated sample after ,, ,, ,, R
(w_3-w_4) = mass of interlayer water after ,, ,, ,, R
(w_4-w_2) = mass of adsorbed water after ,, ,, ,, R

R %	w_3 gram.	w_4 gram.	w_3-w_4 gram.	w_4-w_2 gram.	$(w_3-w_4)/w_3$ %	$(w_4-w_2)/w_3$ %
0	$0{\cdot}431_3$	$0{\cdot}423_0$	$0{\cdot}008_3$	$0{\cdot}003_3$	1·92	0·77
5	$0{\cdot}437_3$	$0{\cdot}430_1$	$0{\cdot}007_2$	$0{\cdot}010_4$	1·65	2·38
10	$0{\cdot}440_1$	$0{\cdot}431_4$	$0{\cdot}008_7$	$0{\cdot}011_7$	1·98	2·66
15	$0{\cdot}445_0$	$0{\cdot}432_6$	$0{\cdot}012_4$	$0{\cdot}012_9$	2·79	2·90
20	$0{\cdot}448_2$	$0{\cdot}433_3$	$0{\cdot}014_9$	$0{\cdot}013_6$	3·32	3·04
25	$0{\cdot}454_0$	$0{\cdot}433_3$	$0{\cdot}020_7$	$0{\cdot}013_6$	4·56	3·00

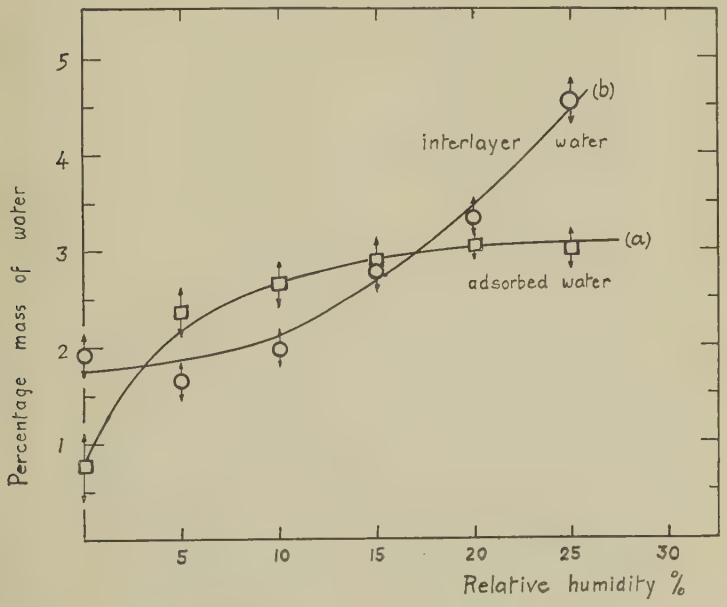

FIG. 1. Adsorbed water, curve (*a*), and interlayer water, curve (*b*), associated with halloysite at various relative humidities; the water masses are expressed as percentages of the mass of mineral, halloysite + water, in equilibrium at each humidity.

rapidly at higher humidities. That curve (*a*) does not diminish exactly to zero at zero humidity may be due to experimental error, but there seems no question that the residual interlayer water shown by curve (*b*) is a real effect, and indeed the X-ray results lead to a similar conclusion. The estimated probable errors arising from the weighings are shown by vertical lines in fig. 1.

Méring (1946) finds a similar curve for the adsorbed water on montmorillonite for the lower range of humidities, though the amount is much greater, being about 10 % at a 20 % humidity compared with 3 % for halloysite. This difference is not surprising since montmorillonite probably possesses a much larger specific surface. The interlayer water curve for montmorillonite shows no residual water at small humidities such as we find for halloysite, but Dr. Méring has pointed out in a private communication that the method used would not enable any residual interlayer water to be measured, but that X-ray measurements, similar to those described in the next section, indicate such residual water in montmorillonite at small humidities.

X-ray study of the dehydration of halloysite.

Specimens prepared for X-ray examination were subjected to various relative humidities at the same time as the experiments already described were in progress. After standing 2–4 days in atmospheres of known humidity, the specimens were quickly sealed between thin mica flakes and transferred directly to an X-ray camera of the Guinier type. In fig. 2 microphotometer traces of the X-ray photographs are shown for the intensity distribution in the first-order basal reflection (001). The second-order reflection is of little use for following the changes which occur since it is unobservable for halloysite, while the third order from halloysite almost coincides with the second order from metahalloysite.

Fig. 2 shows that as the relative humidity is reduced there is a progressive growth of metahalloysite accompanied by a gradual disappearance of halloysite. The lattice spacings of the peaks diminish with the humidity, the halloysite spacing decreasing from about 10·1 to 9·5 Å., and the metahalloysite spacing from about 7·9 to 7·5 Å. From the areas of the component peaks approximate quantitative data can be obtained for the increase of one mineral and the decrease of the other. This entails a consideration of the relative intensities of the two reflections, the details of which are separately discussed in an appendix to the paper. There we show that in a mixture of halloysite and metahalloysite, the

reflected intensity of the (001) reflection per unit mass of halloysite is 4·28 times as great as that of the (001) reflection of metahalloysite.[1] In consequence of this large ratio, the diagrams of fig. 2 at first sight overemphasize the halloysite content compared with the metahalloysite content. There is, for example, about 40 % of metahalloysite in the naturally occurring material, a result which would not be suspected from the appearance of the curve.

In fig. 3 the spacings and percentage amounts (A_H and A_M) of the two minerals are plotted against the relative humidities; the corresponding data are also shown for the original material before treatment. The rapid decay of halloysite and growth of metahalloysite below 30 % humidity are clearly shown. Halloysite which has not changed over to metahalloysite shows a partial shrinkage to a spacing of 9·5 Å., but the process cannot be followed beyond this point. Metahalloysite when first formed has an expanded lattice presumably due to the inclusion of residual water layers which are partially expelled as the humidity decreases to zero. We have been unable to obtain evidence for the existence of intermediate stages with spacings between 9·5 and 7·9 Å. It appears that even at 0 % humidity all the residual water is not removed because the spacing is still markedly greater than that of metahalloysite baked at 300° C., namely, 7·2 Å. This is in general agreement with the results from the weighing experiments.

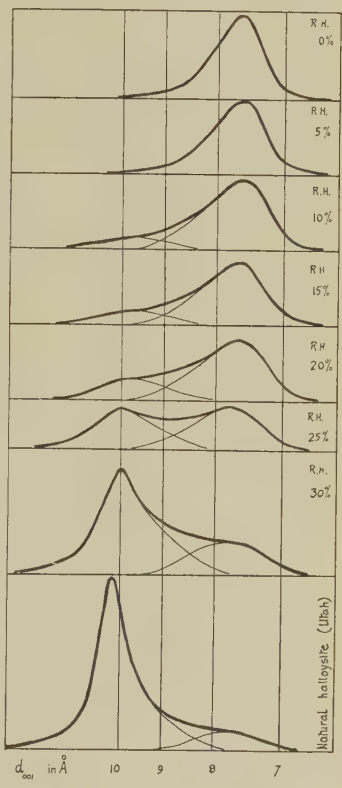

Fig. 2. Microphotometer curves of X-ray powder diagrams of halloysite in equilibrium at various relative humidities. The curves show the (001) reflections from halloysite and metahalloysite. The lowest curve corresponds to the original Utah 'halloysite'.

[1] This value, 4·28, is obtained from comparisons of reflections from specimens having spacings 10·1 and 7·4 Å. Whether we are strictly justified in applying this result when the spacings differ from these values is questionable, but at present there is no obvious alternative.

X-ray diffraction by layer silicate minerals with randomly interposed water layers.

On the basis of a theory developed by Hendricks and Teller (1942) we may estimate the content of interlayer water in halloysite and metahalloysite from their observed lattice spacings. Brindley and Robinson (1946) have already made a simple correlation of the excess

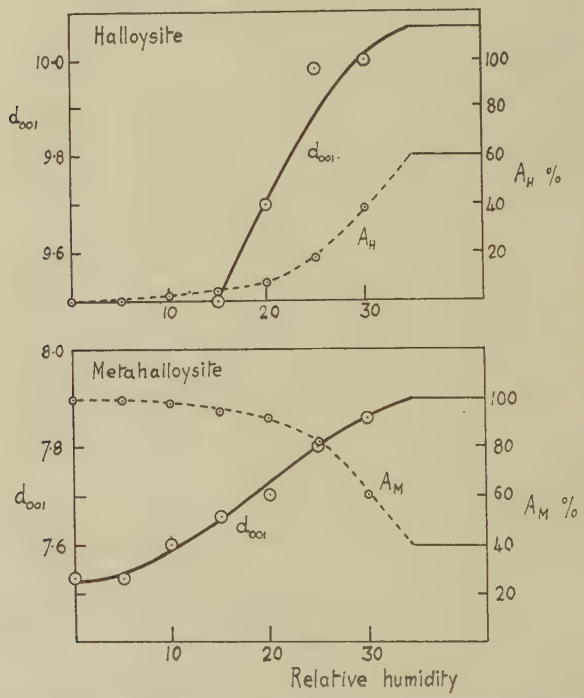

Fig. 3. Basal spacings (full curves) and percentage amounts of halloysite and metahalloysite (dashed curves) in equilibrium at various relative humidities. Horizontal lines at right-hand side give data for the original material which is a mixture of halloysite and metahalloysite.

water found in chemical analyses of metahalloysite with the expanded lattice spacing by calculating the mean lattice spacing resulting from the insertion of a number of water layers randomly amongst the silicate layers. A more satisfactory approach to the problem, however, is on the basis of the Hendricks–Teller theory.

We consider halloysite and metahalloysite to be composed of silicate layers with randomly interposed water layers; when the ratio of silicate layers to water layers, x, tends to unity, the mineral is termed halloysite

and has a spacing of the order of 10 Å., and when x is greater than about 3, the mineral is metahalloysite and has a spacing between about 7·2 and 8 Å. The theory of Hendricks and Teller will be applied in the same simple manner as they themselves have applied it to the corresponding problem for montmorillonite. The randomly interposed water layers create phase shifts between reflections from the silicate layers but do not themselves contribute appreciably to the scattered (or reflected) intensity. There are then two phase shifts to consider between reflections from successive silicate layers, viz.:

$\phi_1 = 4\pi(7{\cdot}13)(\sin\theta)/\lambda$, when there is no interposed water layer[1]

and $\phi_2 = 4\pi(10{\cdot}1)(\sin\theta)/\lambda$, when there is an interposed water layer.

The probabilities p_1 and p_2 of these phase shifts occurring are respectively

$$p_1 = (x-1)/x \quad \text{and} \quad p_2 = 1/x.$$

The theory expresses the average intensity I scattered in a direction θ as

$$I \simeq (1-C^2)/(1-2C\cos\bar{\phi}+C^2), \tag{1}$$

where

$$C = \sum_n p_n \cos(\phi_n - \bar{\phi}), \tag{2}$$

$\bar{\phi}$, denoting an average value, is defined by

$$\sum_n p_n \sin(\phi_n - \bar{\phi}) = 0, \tag{3}$$

and p_n is the probability of a phase shift ϕ_n. For numerical calculations where only two phase shifts are involved, a more convenient expression for I can be developed as follows:

If we put $$p_1\sin\phi_1 + p_2\sin\phi_2 = A$$

and $$p_1\cos\phi_1 + p_2\cos\phi_2 = B,$$

then from equation (3) $\tan\bar{\phi} = A/B$, and from equation (2) we find

$$C = A\sin\bar{\phi} + B\cos\bar{\phi} = B/\cos\bar{\phi}.$$

Substituting in equation (1) we obtain

$$I \simeq (1-A^2-B^2)/(1-2B+A^2+B^2)$$

$$= \frac{(x-1)\{1-\cos(\phi_2-\phi_1)\}}{(x^2-x+1)+(x-1)\cos(\phi_2-\phi_1)-x\{(x-1)\cos\phi_1+\cos\phi_2\}}.$$

This final expression for I, though apparently more complex than equation (1), is now in a form which allows I to be calculated directly as a

[1] 7·13 Å. is the layer spacing of kaolinite and may be taken as the thickness of the kaolinite layers in 'contact' with each other.

function of $(\sin\theta)/\lambda$ for any chosen value of x. Such intensity distributions have been calculated from this expression for a range of values of x from 1 to 20, and the results for the variation of mean lattice spacing with water content are shown graphically in fig. 4.

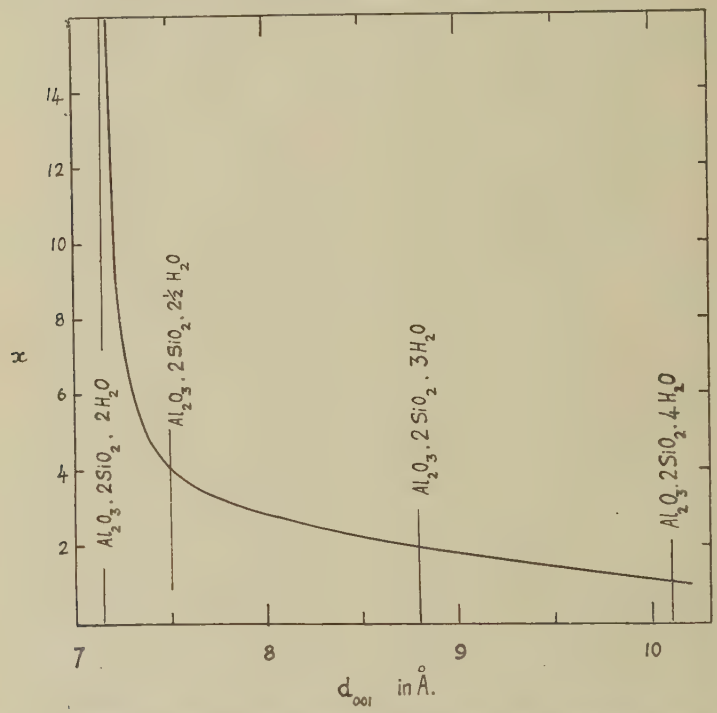

FIG. 4. Variation of mean lattice spacing of halloysite and metahalloysite with water content; x is the ratio of silicate layers to water layers.

The data of fig. 4 may be applied directly to the observed lattice spacings to find the ratio x of silicate to water layers and hence to find the content of interlayer water in the two minerals as a function of the humidity.

Assuming the structure and density of the water layers are as given by Hendricks and Jefferson (1938), i.e., two molecules of H_2O for each kaolinite 'molecule' $Al_2O_3.2SiO_2.2H_2O$, we have

$$\frac{\text{Mass of interlayer water}}{\text{Mass of silicate layers}} = \frac{2\ (\text{Molecular weight of } H_2O)}{x\ (\text{Molecular weight of kaolinite 'molecule'})}$$

$$= \frac{36}{x \times 258}.$$

But the mass of the silicate layers in the 0·5 gram samples used in the dehydration experiments was the quantity designated w_2 which had the value 0·4197 gram (see table I). Hence

$$\text{Mass of interlayer water} = \frac{36 \times 0{\cdot}4197}{x \times 258} = \frac{0{\cdot}0586}{x}.$$

If in a mixture of halloysite and metahalloysite there is a proportion A_H of halloysite with spacing corresponding to x_H, then the interlayer water in the halloysite component is $0{\cdot}0586 A_H/x_H$ and similarly in the metahalloysite component is $0{\cdot}0586 A_M/x_M$. Values of A_H and A_M are obtained from the data of fig. 3, while x_H and x_M are obtained directly from the observed lattice spacings with the aid of fig. 4.

TABLE II. Comparison of interlayer water in halloysite and metahalloysite from X-ray data and from direct weighing experiments.

Relative humidity	Halloysite. d_{001}	Halloysite. x_H	Halloysite. A_H %	Metahalloysite. d_{001}	Metahalloysite. x_M	Metahalloysite. A_M %	Interlayer water in 0·5 gr. sample. in halloysite component.	in meta-halloysite.	Total.	$w_3 - w_4$ table I.
0 %	—	—	—	7·53	4·0	100	—	0·0147	0·0147	$0{\cdot}008_3$
5	—	—	—	7·53	4·0	100	—	0·0147	0·0147	$0{\cdot}007_2$
10	—	—	—	7·60	3·6	98	—	0·0160	0·0160	$0{\cdot}008_7$
15	9·52	1·45	4·7	7·66	3·4	95	0·0019	0·0164	0·0183	$0{\cdot}012_4$
20	9·69	1·32	7·4	7·70	3·3	93	0·0033	0·0165	0·0198	$0{\cdot}014_9$
25	9·98	1·11	18·0	7·80	$3{\cdot}1_5$	82	0·0095	0·0152	0·0247	$0{\cdot}020_7$
30	10·0	1·10	39·0	7·86	3·0	61	0·0208	0·0119	0·0327	—

Hence the total interlayer water in a mixture of the two forms of the mineral can be obtained and the results compared directly with those from the weighing experiments. The numerical data are given in table II and the variations of interlayer water content with humidity obtained by the two methods are compared in fig. 5.

Discussion of results for interlayer water in halloysite and metahalloysite.

Since the errors and assumptions involved in the two methods of finding the interlayer water are wholly unconnected with each other, the large measure of agreement found between the results suggests that a fairly truthful account of the amount of water between the silicate layers in the two forms of halloysite has been obtained.

The X-ray method has the advantages (*a*) of showing the presence of both forms of the mineral for humidities exceeding about 15 %, and (*b*) of enabling the interlayer water in both forms to be determined. Table II shows that x increases from unity to about 1·5 in halloysite before this form completely disappears; $x = 1{\cdot}5$ means that on the

average there are three silicate layers to two water layers, or one out of every three water layers of halloysite has disappeared. The corresponding formula is $Al_2O_3.2SiO_2.3\frac{1}{3}H_2O$. Beyond this stage the water seems to emerge so rapidly that we cannot observe any further stage until we come to a state when x is about 3, i.e. three silicate layers to one water layer, which corresponds to $Al_2O_3.2SiO_2.2\frac{2}{3}H_2O$. The water now emerges

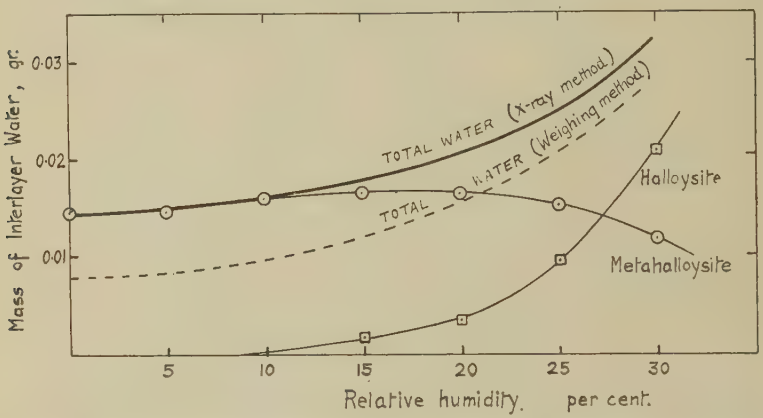

FIG. 5. Mass of interlayer water per half-gram sample of Utah halloysite brought to equilibrium at various relative humidities. Full lines obtained by X-ray measurements of basal spacings, dashed line obtained from direct weighing experiments.

much less easily, and in the driest atmospheres x has not increased beyond about four corresponding to $Al_2O_3.2SiO_2.2\frac{1}{2}H_2O$. These values of x are based on the observed X-ray spacings and the formulae assume the H_2O molecules in the water layers to be packed in the manner described by Hendricks and Jefferson (1938).

Now for humidities less than 15 %, where only metahalloysite occurs, the results from the weighing experiments give for the interlayer water 0·08 gram per half-gram sample (see table I) which corresponds to $x = 6$ and a formula $Al_2O_3.2SiO_2.2\frac{1}{3}H_2O$, a result which is in close agreement with that deduced from the X-ray measurements.

As regards the result that the X-ray measurements indicate rather more interlayer water at the lowest humidities than the direct weighing experiments, the following observations may be made. In the first place, if the water molecules in the residual water layers are less closely packed than in the full hydrated mineral, then the *number* of water layers given by the X-ray measurements would correspond to a *smaller* water content. Secondly, when describing the weighing experiments, we stressed that

if any interlayer water re-entered the sample baked at 300° C., this would lead to an *under*-estimate of the interlayer water. There are therefore good reasons why the X-ray method may give high values and the weighing experiments low values for the interlayer water.

Comparison of present results with published analyses of halloysite and metahalloysite.

It is of interest to compare the present results with various chemical analyses. In a recent discussion of the nomenclature of these minerals, MacEwan (1947) has listed the number of water molecules, n, obtained from a series of analyses published from 1816 onwards. He has rejected data 'for which the SiO_2:Al_2O_3 ratio differed markedly from 2' (p. 39) and has calculated n with reference to one Al_2O_3 group per 'molecule'. The values of n lie in two main groups, (i) between 3·5 and 4·5, (ii) between 2·2 and 2·7, corresponding presumably to (i) halloysite and (ii) metahalloysite with residual water layers. Out of 27 values of n, four only lie between 2·7 and 3·5. There appears to be a definite avoidance of values around $n = 3$. While noting these results, we cannot be very certain of their precise significance. MacEwan describes the analyses as relating to 'air-dry or lightly dried material'. If the material was dried at room-temperature in an atmosphere of 15 % humidity or less, then the present experiments show that it would pass to metahalloysite; we cannot dry halloysite, i.e. remove adsorbed water, without also losing interlayer water. It seems very probable that in all the analyses where n lies in the range 3·5–4·5, the water is partly due to surface adsorption. There is also the question of whether the materials analysed were partly halloysite and partly metahalloysite, as we have found to be the case for the Utah halloysite. For these reasons we consider that, as regards the present work, not much significance can be attached to the amount of water shown in chemical formulae based on early analyses.[1]

The more recent data of Ross and Kerr (1934) are in a different category. Their analyses record H_2O+ and H_2O-, the former presumably being water driven off above 100° C. or thereabouts, though the temperature is not stated. In calculating the number of water molecules, n, corresponding to the H_2O+ content, it is difficult to know whether to refer the calculation to two molecules of SiO_2 or one molecule

[1] This, of course, does not imply any criticism of MacEwan's paper; he was concerned with showing that for a period of a century or more the term 'halloysite' has been used for material containing approximately $2H_2O$ and approximately $4H_2O$ per formula.

of Al_2O_3, because the $SiO_2 : Al_2O_3$ ratio departs from the ideal value 2 rather considerably in some cases. If this is due to an excess of some aluminous material, then the silica-water ratio will be the more reliable basis, but Ross and Kerr could not find, either optically or by X-rays, any impurities to substantiate this view. In table III are listed the values of n calculated from the twelve analyses, nos. 2–13, given by Ross and Kerr based on both the silica and the alumina contents, together with the silica/alumina ratio. The values based on silica are probably the more reliable.

TABLE III. Water molecules per formula, n, corresponding to H_2O+ derived from analyses of metahalloysite by Ross and Kerr (1934), based on (*a*) silica content, (*b*) alumina content.

Specimen no.	2.	3.	4.	5.	6.	7.
SiO_2/Al_2O_3 ...	2·06	1·80	1·95	2·01	1·91	1·93*
n (*a*) ...	2·22	2·64	2·22	2·27	2·23	2·23
n (*b*) ...	2·28	2·38	2·16	2·28	2·13	2·16

Specimen no.	8.	9.	10.	11.	12.	13.
SiO_2/Al_2O_3 ...	1·82*	1·96	1·95	1·70*	1·90*	1·82*
n (*a*) ...	2·35	2·23	2·16	2·93	2·26	2·30
n (*b*) ...	2·14	2·19	2·11	2·50	2·15	2·09

* The values for these ratios given by Ross and Kerr appear to have arithmetical errors: the values given in the table are re-calculated from the published analyses.

For the most part n lies between 2·2 and 2·4, which is entirely in accordance with our own conclusion that for material dried at room-temperature and at humidities less than 10 %, n lies in the range $2\frac{1}{3}$–$2\frac{1}{2}$. As regards the slightly lower values from the data of Ross and Kerr, it must be remembered that their analyses are for material heated (presumably) to 100° C.; data which we give in the following paper, Part III, show that the basal spacing of metahalloysite is appreciably reduced by heating at even so low a temperature as 100° C.; this diminution of spacing indicates a smaller interlayer water content.

We consider, therefore, that our findings are entirely in accordance with the analytical results collected by MacEwan and also with the analyses of Ross and Kerr.

Summary and conclusions.

Two entirely different methods are described for determining the interlayer water content in halloysite and metahalloysite in relation to relative humidity. The first is a direct weighing method in which

samples of natural material and material baked at 300° C. are brought to equilibrium at different humidities. With certain assumptions, which are discussed, it is possible to determine both the adsorbed water and the interlayer water separately. The second method involves measuring the basal spacing of the mineral and applying the Hendricks–Teller theory of X-ray diffraction by a statistical arrangement of water and silicate layers. This method has the advantage of giving separately the interlayer water in halloysite and in metahalloysite when both components are present. The results obtained by the two methods are shown to be in satisfactorily close agreement. It is found that when the natural material is placed in atmospheres of decreasing humidity, not only does the halloysite component change to metahalloysite, but also both components show progressive shrinkage, so that halloysite has a basal 001 spacing in the range 9·5–10·1 Å. and metahalloysite in the range 7·5–7·9 Å. There are corresponding variations in water content, so that in the formula $Al_2O_3.2SiO_2.nH_2O$, n lies between $3\frac{1}{2}$–4 for halloysite and $2\frac{1}{3}$–$2\frac{2}{3}$ for metahalloysite. The fact that $n > 2$ for metahalloysite dried at room-temperature means that it retains an appreciable number of water layers, of the order of one water layer to six silicate layers. These results are shown to be in general agreement with chemical analyses of halloysite collected by MacEwan and of metahalloysite given by Ross and Kerr.

Acknowledgements.—We wish to record our thanks to Mr. Desmaroux, Director of the Laboratoire Central des Services Chimiques de l'État, Paris, for the privilege of working there during the autumn of 1947, where much of the experimental work was done; and especially to Mr. J. Méring and Mlle R. Glaser for their valuable and cordial assistance. We wish to thank also Dr. G. Nagelschmidt for the gift of Utah halloysite, and Professor A. L. Roberts and Dr. R. W. Grimshaw for differential thermal analyses. One of us (J. G.) also wishes to thank the Department of Scientific and Industrial Research for a maintenance grant.

Appendix

Comparison of intensities of (001) reflections from halloysite and metahalloysite.

In order to carry out the investigation described in this paper it was necessary to know the relative intensities of the (001) reflections from these minerals. It was deemed undesirable simply to assume without proof the layer structure of halloysite given by Hendricks in order to

calculate this ratio; conversely, a quantitative measurement of the ratio would provide additional evidence for or against Hendricks's view of the structure. The experimental determination proved more difficult than was anticipated, mainly because the original halloysite already contained a large admixture of metahalloysite. Mixtures of the original material with additional metahalloysite were studied, but the results obtained were very variable. A method involving separate exposures of the original material and of metahalloysite was therefore used (cf. Brindley and Spiers, 1938). A powder holder was constructed containing two flat shallow cavities, each filled with one of the minerals, and the two specimens were brought alternately into the incident X-ray beam, each for a period of five minutes, one specimen reflecting to the right and the other to the left of the beam. The process was continued for a period of about four hours, i.e. two hours for each specimen, while the X-ray tube was operated as steadily as possible in the hope that any fluctuations of X-ray output would cancel out. The two photographs were recorded side by side on a single strip of film so that both had identical photographic treatment. This method has an advantage over a mixture method in that the background scattering from the two substances is not superimposed.

When reflections from two substances, denoted by subscripts 1 and 2 for halloysite and metahalloysite respectively, are recorded in this manner, the ratio of the reflected intensities is given (cf. Brindley and Spiers (1938) p. 29) as:

$$\frac{I_1}{I_2} = \frac{p_1 N_1^2 F_1^2 \phi(\theta_1) A_1 \mu_2 t_1}{p_2 N_2^2 F_2^2 \phi(\theta_2) A_2 \mu_1 t_2},$$

where

$$\phi(\theta) = (1+\cos^2 2\theta)/(\sin\theta \sin 2\theta),$$

$$A = \sin(2\theta-\alpha)/[\sin(2\theta-\alpha)+\sin\alpha],$$

p is the multiplicity factor for a reflection, N the number of unit cells per unit volume of crystalline material, μ the linear adsorption coefficient of the crystalline material (not of the powder specimen), t the exposure time, θ the Bragg angle, and α the angle between the incident X-ray beam and the flat powder surface. In the present case $p_1 = p_2$, $t_1 = t_2$, $N_1/N_2 = 7{\cdot}2/10{\cdot}1$, $\theta_1 = 4^\circ\ 22'$, $\theta_2 = 6^\circ\ 8'$, $\mu_1 = 59{\cdot}4$, $\mu_2 = 79{\cdot}6$ when Cu-$K\alpha$ radiation is used; the angle α was generally taken to be 3°. Assuming the sequence of atomic layers given by Hendricks and Jefferson (1938) and using the atomic F-values of Bragg and West (1928), we find $F_1^2 = 10{\cdot}96 \times 10^3$, $F_2^2 = 3{\cdot}19 \times 10^3$; F_1^2 (i.e. for halloysite) is much greater than F_2^2, mainly because the halloysite reflection occurs at an

appreciably smaller angle. The final calculated value of I_1/I_2 *under the conditions presupposed* is $(I_1/I_2)_{\text{calc}} = \mathbf{4{\cdot}04}$.

The admixture of metahalloysite in the original material complicates the experimental determination of (I_1/I_2). If

$$\text{area of halloysite peak} = H,$$
$$\text{area of metahalloysite impurity peak} = m,$$
$$\text{area of metahalloysite peak from separate mineral} = M,$$
$$\text{and if } I_1 \text{ (halloysite)}/I_2 \text{ (metahalloysite)} = k,$$

then in the absence of any metahalloysite impurity, the halloysite peak would have the area $(H+km)$ and therefore

$$k = (H+km)/M, \quad \text{i.e. } k = H/(M-m) = (I_1/I_2)_{\text{Expt.}}$$

From four separate determinations, we find

$$(I_1/I_2)_{\text{Expt.}} = 3{\cdot}3,\ 3{\cdot}4,\ 3{\cdot}6, \text{ and } 5{\cdot}1$$
$$= \mathbf{3{\cdot}8 \pm 0{\cdot}6}.$$

The observed value is therefore probably somewhat smaller than the calculated value, but the experimental accuracy hardly justifies us attaching any significance to the difference. The experiments have been carried out with all possible care, but there are unavoidable sources of error inherent in such experiments which make the measurement of the intensities subject to considerable uncertainty, namely: (i) the reflections are broad and the peaks do not stand out sharply above the general background; (ii) the reflection from the metahalloysite impurity overlaps considerably that from the halloysite, and in the expression for k, the quantity m is about 40 % of M. The agreement is perhaps as good as can be expected in view of these difficulties. The result, however, tends to support Hendricks's structure for halloysite.

With this justification for the calculated ratio $(F_1/F_2)^2 = 10{\cdot}96/3{\cdot}19$, we now use it to calculate (I_1/I_2) for different experimental arrangements. In the first place, for a mixed powder in which the mass ratio is (g_1/g_2) we have (see Brindley and Spiers (1934); Brindley (1945)):

$$\frac{I_1}{I_2} = \frac{N_1^2 F_1^2 \phi(\theta_1) A_1 g_1 \rho_2}{N_2^2 F_2^2 \phi(\theta)_2 A_2 g_2 \rho_1}$$

when the powder specimen is in the form of a flat block, and ρ_1 and ρ_2 are the densities, respectively 2·10 and 2·59, calculated from the cell dimensions and contents. Then with $\alpha = 3°$, we find

$$\frac{I_1}{I_2} = 3{\cdot}73 \frac{g_1}{g_2}.$$

When photographs are taken by transmission of X-rays through thin layers, as in the dehydration experiments described earlier, then provided the radiation has practically equal paths for the two reflections under consideration—and this condition is amply satisfied in the present investigation—we have

$$\frac{I_1}{I_2} = \frac{N_1^2 F_1^2 \phi(\theta_1) g_1 \rho_2}{N_2^2 F_2^2 \phi(\theta_2) g_2 \rho_1} = 4{\cdot}28 \frac{g_1}{g_2}.$$

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X-ray studies of halloysite and metahalloysite.

Part III. Effect of temperature and pressure on the transition from halloysite to metahalloysite.

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Physics Laboratories, University of Leeds.

[Read November 25, 1948.]

THIS paper concludes the account of our experiments on halloysite and is to be read in conjunction with the two previous parts, to which frequent reference will be made.

Effect of temperature.

The effect of heat-treatment on metahalloysite was examined with the primary object of ascertaining if a recrystallization or 'kaolinization' of metahalloysite could be achieved. The question is of interest in connexion with the possible formation of kaolinite according to the following scheme:

$$\text{silica} + \text{alumina} + \text{water} \rightarrow \text{halloysite} \rightarrow$$
$$\rightarrow \text{metahalloysite} \rightarrow \text{kaolinite}.$$

The experiments, however, provide no evidence for any such reorganization of the silicate layers. The temperature was raised progressively at intervals of about 50° to 450° C., at which temperature the silicate layers commence to decompose, and was maintained for several hours at each temperature.

Attention was directed especially to the appearance of the two-dimensional diffraction bands which would be expected to show some change if reorganization of the layers occurs. This negative result is in accordance with data for other layer-lattice silicates having randomly displaced layers, cf. data for antigorite by Aruja (1945) and by Ali and Brindley (1948) for the transition of pennine into olivine.

The effect of heat-treatment on metahalloysite is also of interest in connexion with the dehydration of the mineral, for, as we have shown in Part II, halloysite is only partially dehydrated in completely dry

atmospheres, and this is also true at low temperatures, such as 100° C. The continued dehydration of the mineral as the temperature is raised is shown by the change of the basal reflections, which become sharper and also change their positions. The (001) spacing diminishes progressively (fig. 1), tending finally to a minimum value of about 7·2 Å., which approximates to the basal spacing of kaolinite, 7·13 Å. As we have shown in Part II, the larger spacing normally found for the (001) reflection, about 7·5 Å., is the result of the inclusion of residual water

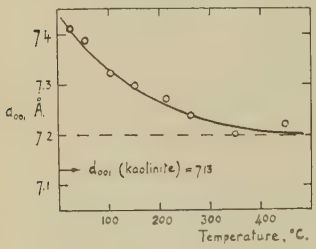

FIG. 1.

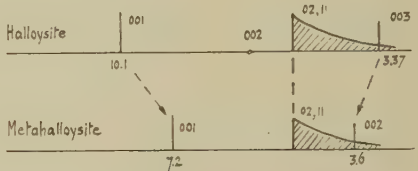

FIG. 2.

FIG. 1. Spacing of (001) reflection from metahalloysite plotted against temperature of heat-treatment. Corresponding value for kaolinite indicated by arrow.
FIG. 2. Diagrammatic representation of the change of the low-order reflections when halloysite dehydrates to metahalloysite.

layers between the silicate layers. It is now seen that considerable heat-treatment is necessary to expel this residual water and, in fact, a temperature so high that the silicate layers themselves begin to decompose is necessary before the water is finally driven out. These results agree with the general picture sketched by Brindley and Robinson (1946) of water inclusions trapped between kaolinite layers. They agree also with the dehydration data of Ross and Kerr (1934, p. 140) who recorded a gradual loss of water on heating halloysite to 400° C., whereas kaolinite loses very little water up to this temperature.

The second-order basal reflection, (002), shows less change than the first order and is less accurately measurable on account of the widely spreading (02,11) diffraction band (cf. fig. 1 of Part I). We find, however, a small *increase* of the (002) spacing accompanying the *decrease* of the (001) spacing, a result which is in agreement with the theory of Hendricks and Teller (1942) as we indicate diagrammatically in fig. 2.

The sharpening of the basal reflections can arise from several causes. By eliminating the residual water inclusions, the heat-treatment produces a more regular lattice, i.e. a lattice in which only one type of layer exists instead of a statistical distribution of two types, and this alone is

sufficient to sharpen the reflections. The silicate layers may also be distorted by the presence of the water inclusions, and the removal of this distortion will also lead to a sharpening of the reflections. The smallness of the crystal size doubtless also contributes to the broadness of the basal reflections. It is scarcely possible to disentangle these combined effects and we can only draw the general conclusion that heat-treatment produces a more regular sequence of basal layers.

Effect of pressure.

The effect of pressure on the halloysite–metahalloysite transition has been examined primarily to determine whether naturally occurring metahalloysite can be produced from halloysite by application of pressure alone. Small samples of 1–2 grams of finely powdered Utah halloysite were placed between flat-faced steel plungers of 1 inch diameter (area = 0·785 sq. in. or 5·06 sq. cm.) and subjected to loads up to 60,000 lb. (27,200 kg.), i.e. pressures up to about 76,000 lb. sq. in. (5000 kg./sq. cm.) by means of hydraulic machines in the Department of Engineering of this University. This maximum pressure corresponds to the static pressure exerted by a column 17,000 metres in height of density 3 and is therefore comparable with or greater than the pressures exerted on natural deposits. In general the pressure was not maintained for longer periods than 24 hours, but in one case the test was continued for 10 days. After compression, the finely powdered material emerged as disks about 2–3 mm. thick which were examined directly by X-ray reflection from the flat faces. The following results were obtained:

(*a*) A partial transformation of halloysite to metahalloysite proceeds more nearly to completion as the pressure and duration of the test are increased. The process cannot be studied in as much detail as the dehydration produced by dry atmospheres because the basal (001) reflection consists of a broad band covering the whole range of spacings from about 10–7 Å., which cannot be resolved clearly into two components. Typical microphotometer curves of the (001) reflection are shown in fig. 3, and separate peaks corresponding to halloysite and metahalloysite have been tentatively sketched by broken lines.

The main conclusion which follows from fig. 3 is that the transformation proceeds further with increasing time and pressure. Curve (*a*), corresponding to the original Utah halloysite, already shows a proportion of metahalloysite (see Part II). Curves (*b*), (*c*), and (*d*) show the effect of increasing the load, while curves (*e*), (*f*), (*g*), (*d*), (*h*), and (*i*) show the effect of increasing the time. While these results do not prove that

metahalloysite is actually produced in nature by application of pressure alone, it seems very probable, when the geological time-factor is taken into consideration, that this must often have occurred.

Other points may be briefly considered. For small loads and times, curves (*b*), (*c*), (*e*), and (*f*) show a very considerable spreading of X-ray scattering towards smaller angles and the X-ray negatives almost suggest

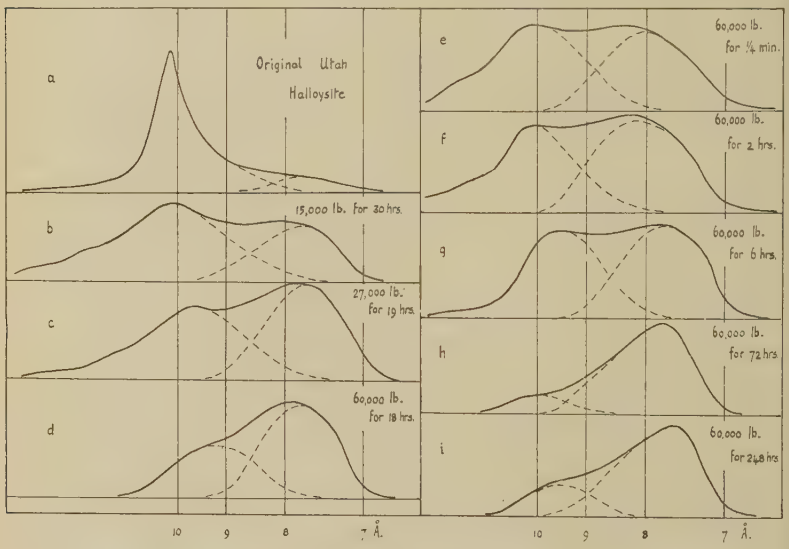

Fig. 3. Microphotometer traces of the (001) X-ray reflection from Utah halloysite before and after application of pressure. A *load* of 60,000 lb. corresponds to a *pressure* of 76,000 lb./sq. in. or 27,200 kg./sq. cm. Vertical lines indicate positions corresponding to lattice spacings of 10, 9, 8, and 7 Å.

the existence of weak broad lines in the range 12–18 Å., which appear in the photometric curves as small variations superimposed on the steady decrease of intensity. We can offer no explanation of these doubtful effects and their reality has not been established for certain. It is also seen that the peak spacings of both the halloysite and the metahalloysite components tend to decrease as the dehydration proceeds, and the process therefore resembles that discussed in Part II. The peak spacings are, however, too inaccurate to justify any detailed discussion.

(*b*) The intensity of the (001) reflection relative to the (02,11) band shows that under pressure a partial orientation of the metahalloysite crystals takes place. This, we believe, is the first time that an orientation

effect has been detected with halloysite or metahalloysite. Kaolinite, on the other hand, shows marked orientation very readily, for example when sedimented in water. Electron microscope pictures of halloysite reveal lath-shaped crystals which produce no obvious orientation when sedimented. When subjected to pressure, however, the (001) reflection is considerably enhanced relative to the (02,11) band. The orientation appears to be confined mainly to the surface layers of the compressed disks. There has previously been some doubt regarding the directions of the crystallographic axes relative to the lath-shaped crystals, but it is clear from this result that the *a*- and *b*-axes must lie in the plane of the lath.

Many natural metahalloysites occur in a hard porcellaneous form and it was thought that these might have been produced naturally by application of pressure. We have examined several such specimens from Lawrence County, Missouri, but without finding any clear evidence for orientation of the crystallites.

Summary and conclusions.

It is shown that heat-treatment up to temperatures at which the silicate layers decompose produces no reorganization of the randomly displaced layers in metahalloysite. No evidence is therefore obtained for the production of kaolinite from halloysite by application of heat. Detailed study of the basal reflections shows that the partial dehydration of halloysite achieved in dry atmospheres and/or at low temperatures can be largely completed by heating to the highest temperature the mineral will withstand without decomposition, about 400° C. The basal spacing then shrinks almost to the value found in kaolinite, and the sharpening of the reflections indicates a more regular sequence of basal layers. Partial transformation of halloysite to metahalloysite has been achieved by application of pressure alone, and probably under geological conditions pressure alone would be sufficient to produce the transformation.

Acknowledgements.—Our indebtedness to those who have kindly supplied us with specimens of halloysite and metahalloysite has already been indicated in Parts I and II. We wish to record our thanks to the Department of Scientific and Industrial Research for a grant to assist a programme of research on clay minerals of which this forms a part and also for a maintenance grant to one of us (J. G.). We thank also Professor R. H. Evans and his assistants in the Engineering Department for their valuable help in carrying out the high-pressure experiments.

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Ships' loadstones.

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[Read June 24, 1948.]

FROM the beginning of classical times it was well known that the mineral conveniently called loadstone, whether magnetite in the strict sense or some other compound, attracted or, better, adhered to iron; also that this property was imparted to a piece of iron in contact with a loadstone, so that a chain of disconnected iron rings could be suspended from it. This action aroused such wonder that many superstitions gathered round the stone which was reckoned throughout the Middle Ages as a gem. A genuine economic value was given to loadstone when the further discovery was made that a suitably shaped piece of the stone, or of iron touched with it, if free to move, would always aline itself approximately north and south. When, where, and by whom this property was discovered and turned to practical use as a direction-indicator, in short the origin of the magnetic compass, has long been a subject of controversy. The evidence cannot here be considered, but it is certain that a primitive compass was familiar in various parts of Europe by A.D. 1200. Informed opinion tends to the conclusion that this knowledge was derived neither from the Chinese nor from the Arabs, but was original to the west. H. Winter (1937) has recently argued, against the claims put forward for Italy and the Mediterranean or for northern France, in favour of a Viking origin. Without further research the only definite evidence is assigned to about the year 1225, somewhat later than the references in the works of Alexander Neckham, the Englishman who was for some time in Paris, and Guyot de Provins; this is a gloss on a passage in the Saga of Floki, the discoverer of Iceland in A.D. 874, as follows: 'in northern lands those who sailed the sea had not then the loadstone.' On the other hand, a northerner who was for many years a superintendent of mines, Emanuel Swedenborg, discusses the question at length (Principia, Cap. XI, Experiment LXXXI) and says that 'the use of the compass at sea seems to have been first known to those inhabitants of Gaul who occupied the shores of the Mediterranean'.

It is possible that in the earliest times the loadstone itself was fixed in a wooden case which would float on water; this method is mentioned

by Petrus Peregrinus, and is reflected in the Italian word for a compass, *bussola*. That there were two poles in a natural magnet had been known since classical times. More often a conveniently shaped piece of iron was touched on the stone to magnetize it and this was floated.[1] Such objects were always to hand on board ship -the needles used for repairing sails; hence the pointer of a compass, though soon made specially and adorned with a fleur-de-lis at the point and made to balance on a pivot, became known generally as a needle. An actual needle magnetically suspended from a loadstone is figured by Gesner (1565). The needle was floated on water either on a straw or stick, the 'festu' of Guyot de Provins (1206), or, according to another poet of about the same date, passed through a cork. Neckham, in the 'De utensilibus' (? 1200) seems to indicate a pivot. A needle attached to a compass-card is regarded as a novelty by Camillus Leonardus in 1502. About the same time Leonardo da Vinci, in an allegorical drawing, shows a wolf steering a boat by means of a compass with a card: by this means deviation from true north, well known by the end of the fifteenth century, could be allowed for.

In earlier times it was generally supposed that the pole of the magnet was influenced by and directed to the north star. This is stated (about 1218) by de Vitry (Hist. of Jerusalem, Cap. LXXXIX), but Roger Bacon (about 1260) doubted it; he demonstrated the property of the 'ugly black stone' to Brunetto Latini (about 1294), whose pupil Dante states that 'the Star the needle's course incites' (Paradiso, XII, 28–31). Another popular theory was that the needle pointed to the original source of the loadstone with which it was touched; North Sea mariners —Norwegians, Danes, French, and English—might for a while have found this idea plausible, as their loadstones mostly came from the extreme north of Europe. Gilbert's book on the magnet, describing his many experiments, appeared in the year 1600. Not everyone accepted his famous conclusion that the earth is itself a magnet: Kircher in 1643 denies it, as does Martin Lister (1698), who not unjustifiably maintains that it is premature to formulate any theory until more is known about the 'effluvia' which pass between the poles of a magnet and the iron, as also that the earth does not as far as is known consist of iron. Gilbert's method of increasing, or concentrating, the power of a loadstone was, however, immediately adopted everywhere. Iron caps or plates were

[1] Michael Scot, probably not later than 1216, says that those who sail the high seas make use of the magnet and iron (L. Thorndike, History of magic and experimental science. New York, 1923, vol. 2, p. 324).

attached at the poles, and a loadstone thus treated was said to be 'armoured' or 'shod'. Kircher figures one corresponding exactly with those now exhibited; Lister mentions one that he was shown in Paris; it 'weighed not above a Drachm and would naked take up a Drachm and a half; but shod it would take up 144 Drachms of Iron'. He adds a caution—'N.B. A strong Loadstone ought to have large Irons, and a weak one but thin Irons; so that a Stone may be over-shod.'

Such mounted loadstones, with the two iron pole-pieces projecting above the body to facilitate application to a needle, were in common use by the middle of the seventeenth century. Webster's 'Metallographia' (1671) says 'like this description are the fragments that we buy at the shops as also that which we have capped with Steel, and bound with Brass or Silver'. Artificial magnets, an easy method of making which was described by John Canton in a paper read in 1750, had probably ousted loadstones by about the end of that century.

Thus for a full 600 years loadstone was an economically important mineral. Pre-eminence over gemstones was claimed for it: Robert Norman, in 'The newe attractive' (1581), described a dispute among precious stones:

> Magnes, the Loadstone I your painted sheaths defie,
> Without my helpe, in Indian Seas the best of you might lye,
> I guide the Pilot's course, his helping hand I am,
> The Mariner delights in me, so doth the Marchant man.
> My vertue lies unknowne, my secrets hidden are,
> By me the Court and Common-weale are pleasured very faire.
> No ship could sayle on seas her course to run aright,
> Nor Compasse shew the ready way, were Magnes not of might.

or Nathaniel Baxter's 'Sir Philip Sydney's Ourania' (1606):

> All mariners of force must be dismayed
> If they should want the Load-stone's blessed ayde;
> By this they saile, else knowe not where they were,
> Such sov'reigntie in Ship doth Magnes beare.

After its supercession for practical use by artificial magnets loadstone degenerated into a mere ore of iron or a scientific curiosity. As late, however, as 1819 Sir Richard Colt-Hoare writes that in Elba at Punta della Calamita, the headland so called from the loadstone rocks there, 'large veins of this substance are intermixed with iron ore'; so some trade may have survived.

The mounted specimens are commonly called 'ships' loadstones' and were sometimes at any rate carried on board ship. Captain Thomas James made a voyage to discover the north-west passage in the years

1631–32; the inventory of the instruments taken includes, besides dozens of 'meridian compasses' and needles, 'four special needles . . . of six Inches Diameter and toucht curiously with the best Load-Stone in England' and 'a Load-Stone to refresh any of these, if occasion were; whose Poles were marked for fear of mistaking'. Again, on a voyage of de Castro in the east in 1538–39 'the stone now used by the pilot to remagnetise the needle was from the coast of India'.

Such loadstones would be used by all compass-makers and by some land-surveyors and travellers, as well as by sea-captains: Pero Tafur in 1439 says, of the Egyptian desert, 'they navigate here with the compass as at sea'. Mine-surveyors seem to have made comparatively little use of loadstones: Agricola (1556) in Book III of the 'De re metallica' gives an elaborate description, with a figure, of compass-cards—'within the innermost circle it is hollowed out to contain a magnetic needle'. The notes and appendix in the Hoover translation mention an earlier work, the 'Nützlich Bergbüchlin' of Calbus (von Kalbe), a mine-surveyor and burgomaster of Freiberg, who died in 1523; this describes and figures a miner's compass, but 'there is not the slightest reference to its use for anything but surface direction of veins'. Porta (1558), who devotes the whole of Book VII of his 'Magia naturalis' to the magnet, also mentions the use for this purpose and 'especially in making tunnels for aqueducts' (Cap. xxvii).

The method of touching the iron with the stone is described in full by Martin Cortes in 1551 and by William Barlowe in 'The navigator's supply' (1597). By this time the single needle had been replaced by two wires, their ends soldered together and their middle parts attached to and separated by the 'fly' or socket-piece to be placed on a pivot. (The Oxford Dictionary gives the word fly as 'a compass card', with the date 1571 for its earliest use.) Barlowe writes:

'As for the touching of the wyars of the Flie with the Load-stone, I would wish it to be performed after this sort. First of all, have a great care of the goodnesse, the quantitie and the forme of the Stone; for if he be never so good, and very small therewith, he can give but small force unto the Compass: and againe, though he be never so great, yet if he be of base qualitie, his touche can be but faint. An ovalle forme, or somewhat longer, retaining a like proportion from the middle to each end, is very good. Alwayes provided that the length of the stone lie according to his owne line of North and South: for a stone of this forme giveth foorth his vertue in the touche a great deale more forcibly then it can, if by reason of the evil shape thereof, his force in himselfe be confusedly dispersed, and not joyntly directed to his due points. The wyers before they bee touched, ought to be polished and made very cleane, and fitted unto the Flie, and then touched after this manner. With the North end of the Stone, presse each wyer, beginning at the middle, and so along unto the ende, that you would have turne Southerly: and with the South end of the

Stone doe the like from the middle of eache wyer, unto the end that you would have turne Northerly. Then glewe them in such sort, that the Flie may stand equally upon his pinne, having both the North and South endes of the wyers uncovered, that their Touche may be refreshed, as occasion shall require.'

Loadstone is said by Pliny to have been found first on Mt. Ida; also at two of the ancient places called Magnesia (in Macedonia and Asia Minor respectively), Cantabria in Spain, Boeotia, the Troad, and Aethiopia (XXXIV, 42 and XXXVI, 25). Isidorus (early seventh century) practically quotes Pliny, but substitutes India for Ida (XVI, iv, 1). India became the best-known locality; it is given by Marbodus (*c.* 1080) and all his imitators, and by de Vitry (Hist. of Jerusalem, LXXXIX) about 1218. Marbodus also mentions the country of the Troglodites, probably equivalent to Pliny's Aethiopia. Albertus Magnus, writing about 1250, adds the interesting fact that he himself saw it mined in Germany (De Min., Lib. II, Tract II, Cap. 11). He does not mention the directive property, but in 1269 Petrus Peregrinus wrote the first treatise specifically on the loadstone. He describes clearly a method of finding the poles and says that the stone is found in northern regions and is brought by sailors to the ports of Normandy, Picardy, and Flanders. Only his epistle on the magnet appears to have survived, but Roger Bacon (Op. Tert., Cap. XIII) declares that he 'knows by experiment all natural history, and physic, and alchemy, and all things in the heavens and beneath them. . . . Wherefore he has inquired into all the operations of metal-founding, and the working of gold and silver and other metals, and of all minerals' (trans. S. P. Thompson). Though I have not found a definite statement earlier than Porta (VII, Cap. vii) in 1558, it seems likely that the loadstone of Elba was known by this time; the exact locality, Cape Calamita, is named from it. The name *calamita* for loadstone, current all round the Mediterranean, is, according to C. H. Haskins, used by Michael Scot about 1217; it is almost certainly derived from Arabic and has been attributed to Rhazes (first half tenth century); Scot, at the court of Frederick II, Stupor Mundi, in Sicily, was one of the earliest scientists to introduce oriental learning to the west. Popular attempts to derive the word *calamita* from Latin or Greek result in nonsense.

The first independent and reliable list of localities for loadstone is given by Agricola in 1546 (De Nat. Foss., V): the Cantabrian province of Spain; an island in the north off the coast of Lapland, which takes its name from Magnes; in Germany he gives, obviously from personal observation, the Harz Forest, Swarzburg, Joachimsthal, Franconia, and Bohemia; for several of these his localities are precise; for example, the

first is a single shaft beyond Harzburg by the seventh (mile)-stone from Goslar, and for others a particular mine is named. He then gives a version of Pliny's list with additional comments—India, near the river Indus and in certain coastal rocks. Agricola evidently was as careful with his references as with his observations, for he points out that Pliny follows, as he himself mentions, an otherwise unknown author, Sotacus. Throughout the seventeenth century others quoted or misquoted, with or without acknowledgement, from Agricola; one Italian writer (Moscado) omits several words and, more disastrously, a semicolon, with the misleading result that he implies an island called Calamita off the Cantabrian coast of Spain.

There is no need to mention here the gradual accumulation of known localities for loadstone, but those in Britain may be of interest; apparently only those in Devon were actually utilized. Unfortunately the earliest records are open to question: Gilbert (Book I, Cap. 2) says 'In England quite lately a huge power of it was discovered in a mine belonging to Adrian Gilbert, gentleman; also in Devonshire and the Forest of Dean' (translation, p. 11). Silvanus Thompson has a note that 'Adrian Gylbert of Sandridge in the Countie of Devon, gentleman, received from Queen Elizabeth a patent for the discovery of a North West passage to China'. He would therefore be interested in loadstones; but Sandridge Park is about four miles west of Brixham. Both that district and the Forest of Dean are well known for deposits of haematite, but not of magnetite. We can only speculate on the explanation; neither locality is mentioned again. Camden in 1607 mentions 'Dertmore where loadstones have lately been found' (Gibson's translation). Childrey's 'Britannia Baconica' (1661) adds that they are 'of good value and vertue'; in the same year Webster (Metallographia, p. 350) says 'I have had it from some Gentlemen of very great worth that lately there is found good store of it in the aforesaid county of very excellent force and vertue'; but Christopher Merrett (Pinax, 1667), after commending the Dartmoor loadstone, adds 'et alibi viliores'. The Philosophical Transactions for 1666 (no. 23) record that a loadstone from Devon weighing 60 lb. was exhibited to the Royal Society; it was not very strong in lift, but moved a needle at a distance of nine feet. Polwhele's 'History of Devonshire' (1797, vol. 1), quoting various topographical writers, mentions the following Devon localities: Sotwardstone, South Brent, North Molton, Ashburton, and Hennock, but adds significantly that the stones from the Persian Gulf are better. John Woodward's collection included two specimens of loadstone from Devon. The only other

mention of British loadstone that I have noticed is Sibbald's remark (1684, Lib. IV, Cap. v) that it is said to occur (not necessarily worked) 'in Rupe Arcis Britannodunensis', i.e. in the Castle Rock of Dumbarton.

Besides the two mounted loadstones exhibited, that in brass belonging to, that in silver on loan to, the Geological Survey Museum, Commander A. M. Hughes, R.N., kindly supplied me with a list of nine in the National Maritime Museum, Greenwich; the earliest, in brass with poles marked N. and S., is attributed to the second half of the sixteenth century; five, in brass, bronze, or silver, to the seventeenth; and three to the eighteenth. In the Museum of the History of Science at Oxford some half-dozen examples include a very small one in gold and one in silver closely resembling that lent to the Geological Survey Museum; this bears an inscription in Russian and the date 1741.

One of the many superstitions regarding the loadstone may be mentioned as it is relevant to the use for compasses. It was held for many centuries that garlick or onion nullified the power of a magnet. I can offer no suggestion as to the origin of this idea. It was refuted by Porta (Magia naturalis, VII, Cap. xlviii) about 1530; confirmed by Van Helmont in 1621; again refuted by Sir Thomas Browne with many experiments. Several other writers have pronounced against it, but such well-established traditions die hard. In the sixteenth and seventeenth centuries there was current definite rule that ships' captains must not eat garlic at sea, lest their breath subsequently cause the compass to deviate and so lead to the loss of their vessels. Perhaps Mediterranean captains carried mounted loadstones to revive the needle if or when they yielded to temptation. The passage in Porta is worth quoting. I have not seen the English translation of 1658, but copy it from Silvanus Thompson's notes:

'It is a common Opinion amongst Sea-men, That Onions and Garlick are at odds with the Loadstone: and Steers-men, and such as tend the Mariners Card are forbid to eat Onions or Garlick, lest they make the Index of the Poles drunk. But when I tried all these things, I found them to be false: for not only breathing and belching upon the Loadstone after eating of Garlick did not stop its vertues: but, when it was all anoynted over with the juice of Garlick it did perform its office as well as if it had never been touched with it. . . . And again when I enquired of Marines, whether it were so, that they were forbid to eat Onions and Garlick for that reason, they said They were old Wives fables, and things ridiculous; and that Sea-men would sooner lose their lives then abstain from eating Onyons and Garlick.'

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(Includes text of Neckham's De utensilibus and of Guyot de Provins's poem La Bible, with notes.)

I am indebted for the reference to Winter to Commander A. M. Hughes, R.N., of the National Museum, Greenwich, and through him to Commander W. E. May, R.N., of the Admiralty Compass Observatory, Slough, for those to Barlowe and De Castro.

A refractive index comparator for the microscope.

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[Exhibited March 10, 1938.]

THIS stage-attachment simplifies microscopic comparison of refractive indices of small transparent grains by the immersion-liquid methods of O. Maschke,[1] Schroeder van der Kolk,[2] and other workers. It was designed in 1938 for use in mineralogy and petrology, but has applications in several branches of pure and applied chemistry, particularly for determining the relative purities of numerous raw materials and refined products of the chemical industry and in checking the constituents of glasses, slags, refractories, and cements. It can be used with advantage, also, in the optical study of soils.

The device enables the composition of a suitable liquid to be changed by small increments of any desired magnitude between certain limits, in a nearly closed transparent vessel under the microscope, until there is little or no significant difference between the refractive index of this liquid and either *the* refractive index, or *a* refractive index, of grains immersed in it, as judged by the Becke method of central illumination[3] or the van der Kolk method of oblique illumination.[4] A suitable refractometer is then used for finding the refractive index of this final liquid. With due care the value of the refractive index so derived can have an accuracy of about $\pm 0{\cdot}002$, or better, if monochromatic light is used throughout and appropriate temperature corrections are applied.

The aim of the instrument is to increase the rate and convenience with which immersion-liquid refractive index comparisons can be made by providing simpler and more efficient means for mixing the necessary liquids than is involved in the method of stirring together drops of liquids of known refractive indices on a plane glass slip, or in a small

[1] O. Maschke, Pogg. Ann., 1872, vol. 145, pp. 565–568; Niedemann's Ann. Physik, 1880, vol. 11, pp. 722–734.

[2] J. L. C. Schroeder van der Kolk, Kurze Anleitung zur mikroskopischen Krystallbestimmung. Wiesbaden, 1898, pp. 11–14. Tabellen zur mikroskopischen Bestimmung der Mineralien nach ihren Brechungsindex. Wiesbaden, 2 Aufl., 1906.

[3] F. Becke, Sitz.-ber. Akad. Wiss. Wien, 1893, vol. 102, Abt. I, pp. 358–378.

[4] J. L. C. Schroeder van der Kolk, Zeits. wiss. Mikroskopie, 1892, vol. 8, pp. 456–458.

glass cell. More especially it enables the whole process of comparison to be carried out, step by step, in a single dust-free container, to be observed continuously under the microscope, and to be controlled closely throughout. Further, at the end of this process of comparison any tendency for a slight change to occur in the refractive index of the final mixture of liquids, due to differential evaporation, is minimized by the fact that the main vessel is sealed except for a pin-hole at one point.

The material and form of the instrument as a whole give a combination of lightness with the necessary mechanical strength (fig. 1). Further, its size is such that it can be accommodated on the stage and rotated through 360° about the objective of any modern petrographic microscope (fig. 2). It is noteworthy that, without losing these advantages, the instrument could be made nearly half as large again than it is at present, thereby increasing the range of certain of its capabilities. The highest power of objective which can be used in conjunction with the device is determined by the fact that a minimum separation of 3·5 mm. must be maintained between the lower surface of this objective and the upper surface of the microscope stage if the objective is to be prevented from touching the liquid mixing-chamber of the instrument. The shape and internal dimensions of the main vessel have been so chosen that clogging by wedged particles of test material is unlikely unless the particles are more than 1·5 mm. in maximum diameter. Finally, appropriate parts of the device can be assembled, filled, emptied, separated, and cleaned with ease and rapidity.

In use, the two most important advantages of the instrument are: (i) that it enables the desired refractive index comparison to be made as a simple progressive operation not involving, for example, repeated transfer of a minute grain, or grains, from a volume of one liquid to that of another, with consequent manipulative and optical complications; and (ii) that relatively small total volumes of immersion liquids (less than 0·6 c.c.) can be employed while retaining adequate accuracy in the values of refractive indices so obtained. Additional optical advantages are that any desired grains can be centred in the field of view and then rotated completely on the microscope stage, and that a platy particle can be tilted and held thus (on its edge, if desired), for microscopic study, by means of controlled air bubbles in the immersion liquid, as is described below.

Though it involves an immersion liquid mixing-chamber comparable with the open glass cell employed by some workers, this instrument differs from the latter in being a nearly closed main vessel carrying a

small detachable reservoir from which drops of a suitable immersion liquid can be introduced into the main vessel at will, and in using a pneumatic method for thoroughly mixing these drops, one or more at a

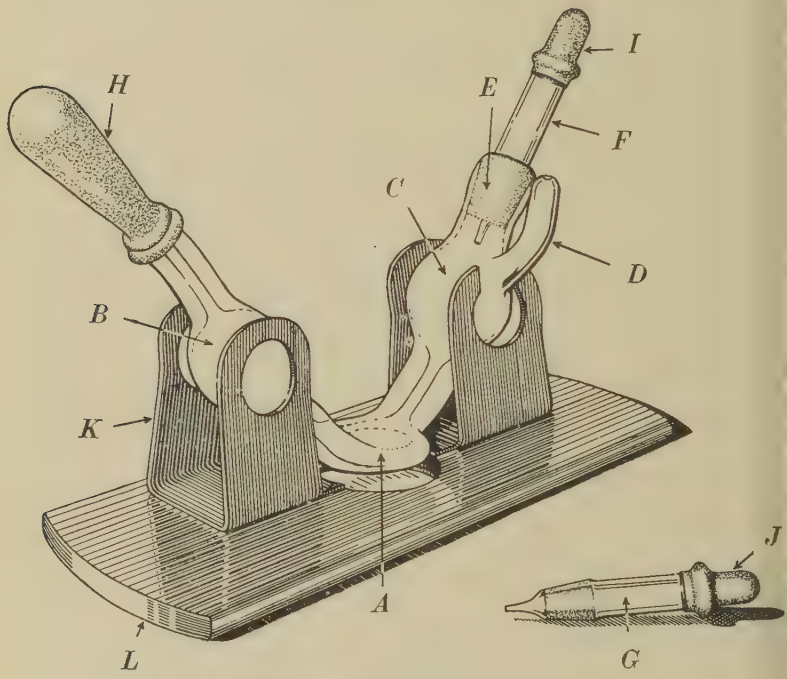

Fig. 1. Sketch of the apparatus.

time, with a small volume of another immersion liquid which has been placed in the main vessel beforehand.

Construction.—From fig. 1 it will be apparent that the main vessel consists of a V-shaped length of pyrex glass capillary tubing with inclined limbs of nearly equal length, bearing a small flattened bulb *A* at the base of the 'V' and a nearly spherical bulb, *B* or *C*, at the centre of each inclined limb. Into a ground-glass joint *E* in the slightly expanded upper end of one of these limbs fits either of two identical cylindrical glass reservoir tubes, *F*, *G*, each with a tapered cylindrical nozzle, while over the flanged upper end of the other limb fits an elongate rubber bulb *H*, such as is used for operating tubular glass fountain-pen fillers. Miniature equivalents (*I*, *J*) of bulb *H* close the respective ends of tubes *F* and *G* which are remote from their nozzles.

Glass bulb *A* is the chamber in which part of the mixing of immersion liquids occurs and where subsequent refractive index comparisons are made. Its upper and lower walls are parallel and have a thickness of about 0·5 mm. The external depth of the chamber is 2·5 mm.

The glass bulbs *B* and *C* serve not only as terminal reservoirs and mixing-chambers for immersion liquids but also as retarding devices to check any tendency for a rapid rise of these liquids into contact with the nozzle of either tube *F* or tube *G* (whichever is being used in the instrument at the time), or with the rubber of bulb *H*. Bulb *C* has a short cylindrical branch tube *D*, fused into it at a point on the surface of the bulb near its upper junction with the main tube. This branch tube is curved upwards, away from the bulb, into approximate parallelism with the main tube and its upper end is level with the top edge of the latter. Here *D* is closed except for a circular hole, nearly three-quarters of a millimetre in diameter, which allows air to enter or leave bulb *C* and its associated parts of the main tube when an immersion liquid is withdrawn from or forced into bulb *C* by manipulation of rubber bulb *H*, but minimizes the chance of loss of any such liquid that might rise high in bulb *C*. Further, evaporation through such a vent at normal laboratory temperatures is so small as to be negligible for any but long periods of time. This means that over the usual duration of a laboratory determination, changes in the refractive index of an immersion liquid in the main tube due to differential evaporation and loss of a more volatile component from a less volatile one can be neglected in practice.

Each of the identical cylindrical glass reservoir tubes *F*, *G* has at one end a smooth-edged, cylindrical nozzle, 0·9 mm. in external diameter, carefully shaped to give small drops of refractive index liquid, while at the other end is a miniature rubber bulb, *I* or *J*, for expelling these drops. Such a bulb is made from the closed end of a fountain-pen ink-sack. On the exterior of the tube immediately above the nozzle is a conical surface which has been ground to make a very close union with a complementary ground surface *E* in the main tube. The uniformity of drops generated from these nozzles must be preserved by keeping the latter scrupulously clean and not allowing them to become chipped as a result of careless handling.

To ensure rapid recovery of form after compression, bulb *H* is composed of heavy resilient rubber, while its open end is of such a diameter and so thickened that it makes a thoroughly airtight union with the surface of the main tube. Care must be taken to prevent any of the immersion liquids which react with rubber from reaching the interior

of this bulb or the refractive index of the liquid in the main tube may be changed by admixture with such solutions and the walls of the bulb caused to stick together when the latter is compressed between the fingers.

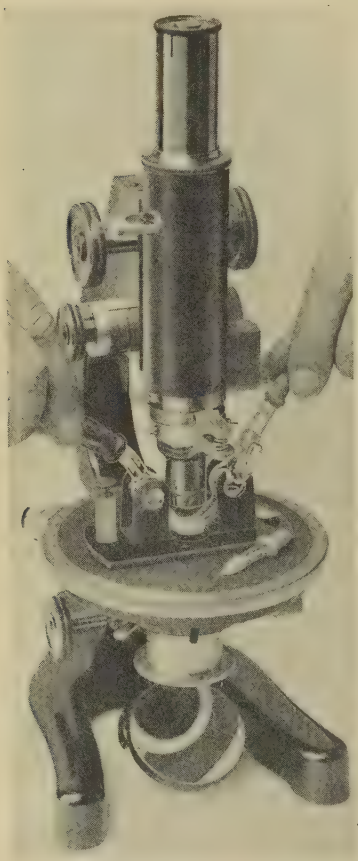

Fig. 2. The apparatus in operating position on the microscope stage, with the bulbs and reservoir tube partly filled with liquid.

The phosphor bronze spring-clip *K* has two pairs of nearly vertical members with circular holes cut in them of a size such that they receive and grip the nearly spherical glass bulbs *B* and *C* when the main tube is pressed down between these members. One of the four members is modified to accommodate the small vent-tube *D* projecting from bulb *C*. This simple spring-clip serves not only as a convenient holder for the main tube, accepting or releasing it in a moment, but also acts as a double ball-pivot which allows the main tube to be swung through 5° either side of the vertical position about a horizontal axis joining the centres of bulbs *B* and *C*. Among other advantages this last possibility enables grains of material under examination in bulb *A* to be segregated, by suitable tilting of the main tube, in a part of the bulb where they will not be swept into the main stream of immersion liquids which are circulated through *A* during the mixing of these liquids.

The base plate *L* of the whole device is a small rectangular strip of stiff bakelite fibre, little more than twice the thickness of most glass specimen slides. It has a central hole through which light reaches bulb *A* from below when the instrument is attached to a microscope. This plate is held in position on the microscope stage by the usual pair of clips provided for holding specimen slides (fig. 2). Consequences of

this mode of attachment are that the contents of bulb *A* can be explored, or any desired grain centred for optical tests, by appropriate small movements of *L* under the slide-clips. While using the instrument, the microscope stage should be maintained in the horizontal position throughout.

Operation.—As a preliminary to the refractive index determination of constituents in a solid sample a representative piece is crushed, but not ground, and then screened to provide particles preferably between 0·5 and 1 mm. in diameter. Should it be necessary, the instrument can accommodate particles up to 1·5 mm. in diameter. If the sample is a detrital sediment or other type of granular material, any initial treatment must be followed by screening, to remove grains greater than 1·5 mm. in diameter.

This is followed, if necessary, by a determination of the approximate refractive index or refractive indices of grains of this screened material, made with a set of immersion liquids differing in refringence by, say, 0·01.

A small amount of the screened material is then introduced into the lower part of bulb *B* with a spatula made from a short length of heavy-gauge platinum wire[1] flattened into a minute blade at one end, and from here the sample is introduced into bulb *A* by suitable tapping.

Any grains remaining in *B* are washed into *A* during the subsequent addition of a definite number of drops of immersion liquid *x* (say, of a refractive index *lower* than that of the sample) from reservoir tube *G*.

The body of liquid resulting from the coalescence of these drops may fill part or all of bulb *A* and more or less of the main tube connecting *A* with *B*. If this last is the case, to prevent accumulation of liquid in the tube when further drops of liquid are added to bulb *B*, air is displaced from bulb *H* by squeezing the bulb—which air passes through *A* into *C* in the form of bubbles and thereby allows the level of liquid to fall in the tube connecting *A* with *B* and rise proportionately in the tube connecting *A* with *C*.

Reservoir tube *F* is nearly filled with immersion liquid *y* (which has a *higher* refractive index than that of the sample and is miscible with immersion liquid *x* in all proportions) and carefully inserted in ground joint *E*.

The main tube is then pressed into spring-clip *K* and the whole device placed on the stage of a petrographic microscope with the slide-clips of the latter gripping the base plate *L*. The stage is then set horizontally

[1] No. 19 S.W.G. is suitable, and the length may be fused into one end of a small glass rod for greater manipulative convenience.

and the apparatus manipulated under the clips to centre any required grain, or group of grains, in the field of view.

A drop of liquid y is expelled from F into the lower part of bulb C by means of the left hand, after which gentle pressure on bulb H between finger and thumb of the right hand causes a column of the liquid x to rise into the lower part of bulb C, followed by a train of bubbles of air from bulb H, thereby mixing this liquid to some extent with the drop of liquid y. On relaxing the pressure on H, the still relatively inhomogeneous mixture of x and y retreats into and through A, followed by a series of air bubbles drawn from bulb C, which bubbles thoroughly agitate and stir the mixture. On repeating this oscillatory circulation of the mixture and bubbles four or five times in rapid succession, by alternate increases and decreases of pressure on bulb H, the essential optical homogeneity of the mixture is attained very rapidly and conveniently, after which a refractive index comparison can be made between it and a chosen grain by either the method of central illumination or that of oblique illumination, using monochromatic light.

Thereafter additional drops of liquid y are expelled from F, one at a time, each mixed in the manner already described, and a corresponding refractive index comparison made with the grain or grains at each step until optical match is obtained. Should one or more drops of y be added in excess of what is required for match, reservoir tube F is replaced by reservoir tube G, containing liquid x, and from it is added to the mixture of liquids the necessary number of drops to rectify the error.

If, during the mixing process described, grains tend to be swept from bulb A into either bulb B or bulb C, or both, by the stream of immersion liquid, the main tube can be swung in its spring-clip through approximately 5° either side of its normal vertical position about an axis joining the centres of bulbs B and C, so that test grains fall, or rise,[1] to the side of A remote from the main tube, i.e. into a 'backwater' out of the main stream of liquid, until the mixing process is completed.

By suitable manipulation of bulb H, trains of a few large air bubbles (or a larger number of small air bubbles) can be induced in the main column of liquid. With a little practice such bubbles can be used to tilt and hold platy grains on edge, for optical examination, aided by a slight cant of the main tube in its spring-clip to help to introduce such grains into the bubble-train which may then be caused to circulate round

[1] Depending upon the direction of tilt of the main tube and upon whether the specific gravities of the grains are greater or less than the specific gravity of the immersion liquid.

only one side of bulb A. In use this technique is as simple and effective as that described by E. S. Larsen[1] for turning small mineral plates in a relatively viscous immersion liquid to study their optics.

As soon as the final liquid mixture is obtained the whole device is removed from the microscope stage, tube F is withdrawn, and a drop of the liquid expelled onto a refractometer prism for direct refractive index measurement at a known temperature, using monochromatic light. If it is not convenient to make this measurement immediately, the final mixture of x and y can be left in the instrument for a considerable period of time without fear of any significant change of composition and therefore of refractive index, due to differential evaporation, provided F is left in position throughout, or replaced by a small well-fitting ground-glass stopper immediately the matching process is complete.

After use the main tube and reservoir tubes of the instrument should be cleaned with benzene, or some other solvent appropriate to the immersion liquid employed, using bulb H for circulating the solvent thoroughly in the main tube, but carefully preventing the solvent from wetting this bulb.

Utility.—Experience gained with this apparatus over the past ten years has shown that it is especially suitable for refractive index determinations on small detrital particles, crushed fragments of minerals, and such microscopic crystals and crystalline aggregates as occur in some soils and in a number of raw and manufactured industrial materials.

Most commonly the device serves as a convenient aid in measuring *one* refractive index of a given material, but *two* or *three* refractive indices of the same material, or of an association of different materials, can be determined successively and during one and the same main operation if the greatest numerical difference between the refringences concerned is sufficiently small and appropriate immersion liquids are employed.

From a relationship discussed by C. Christiansen[2] it can be shown that, for light of a chosen wave-length, and at a given temperature, the refractive index n of a mixture of two immersion liquids x and y is given by the expression:

$$n = (v_x n_x + v_y n_y)/(v_x + v_y),$$

where v_x, v_y are the volumes of liquids having refractive indices n_x and

[1] E. S. Larsen, Bull. U.S. Geol. Surv., 1921, no. 679, pp. 22–23.

[2] C. Christiansen, Niedemann's Ann. Physik, 1884, vol. 22, p. 298; 1885, vol. 24, p. 439.

n_y respectively, for light of the chosen wave-length and at the temperature concerned.

In the apparatus here described the total effective volume of either reservoir tube is 0·15 c.c., and that of the main tube is 0·45 c.c., i.e. a volume ratio of 1:3 is afforded. The significant unit of volume, however, is not the cubic centimetre but the *drop* of any given immersion liquid which is obtainable from the nozzle of either *F* or *G*, since this determines the magnitude of each successive change of refractive index that can be produced when two immersion liquids of different refractive indices are mixed in the instrument. From a clean smooth nozzle of the form shown in fig. 1, and for a given gaseous environment, the volumes of successive drops of a chosen liquid are sensibly equal, but differ in this respect from one liquid to another, since at a given temperature the volumes are functions of the surface tensions of the liquids concerned.[1]

Experiment shows that an average of 12 drops of liquid can be obtained from a single filling of either reservoir tube and therefore a maximum of about 36 drops can be accommodated in the main tube of the device. This gives possible extreme ratios $v_x : v_y = 1:35$ or $35:1$, and proportionate values of n.

These are not the limiting ratios obtainable, however, since the drops discussed hitherto are what may be termed 'full' drops, which form and fall naturally from the nozzle of *F* or *G* on compressing bulb *I* or *J* sufficiently, whereas drops of about *half* this volume, or less, can be derived from the same nozzles by expressing the necessarily smaller amounts of liquid and then causing the hanging droplets to separate and fall to the bottom of bulb *C* by a light tap with the finger-nail, or a pencil, on the outside of ground joint *E*. Such 'half-drops' can change the ratio $v_x : v_y$ to extreme values of the order 1:70 or 70:1, thereby reducing the effective magnitude of the change of n in proportion. It will be evident that such half-drops, or still smaller drops, need to be employed only when the refractive index of a liquid mixture and that of its immersed grains are very nearly equal.

The author is grateful to the late Mr. F. Niedergersass for shaping the necessary glass parts of the first model of this apparatus, and to Mr. A. N. Lanham, chief laboratory assistant in the Department of Mineralogy and Petrology, University of Cambridge, for making its base plate.

[1] W. D. Harkins and F. E. Brown, Journ. Amer. Chem. Soc., 1919, vol. 41, pp. 499–524.

On the delimitation of diorite and gabbro and related rocks.

(With Plate XXV.)

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THERE is general agreement as to the basis on which most major igneous rock-types are defined despite some difference of opinion as to quantitative limits. This is not true of diorites and gabbros (or of their hypabyssal and volcanic equivalents) in distinguishing between which at least four different criteria are in current use.

At the beginning of the microscope era, Zirkel (1866) and Rosenbusch (1877) defined diorite and gabbro as plagioclase-felspar-rocks with hornblende and/or biotite, and with pyroxene, respectively, and this criterion is still used in Harker's 'Petrology for students'. Because the more basic hornblende-diorites, so defined, are chemically indistinguishable from gabbros, and because their hornblende is often altered pyroxene, Rosenbusch (1907, p. 263) later re-defined diorite and gabbro on the basis of their plagioclase-felspars, that of diorites having less, and that of gabbros more, than 50 % of anorthite. It is doubtful whether he meant this distinction to be rigidly applied, but subsequent authorities basing their definitions of types on the composition of the felspar (e.g. Johannsen, Holmes, and Niggli) have so used it. Tröger (1935) created a group of gabbro-diorites for rocks characterized by andesine-labradorite; this corresponds to the belugite family of Spurr (1898).

A number of writers have recently abandoned the felspar criterion in favour of one based on colour-index, probably because (*a*) in the andesites much of the felspar is often occult in the residual glass, and is more sodic than that actually developed, so that the average composition of the felspar cannot be modally determined, and (*b*) because the darker diorites, so defined as having oligoclase or andesine as dominant felspar, are chemically gabbroic (hence Barrell's dictum, quoted by Shand (1927, p. 184), that no rock with a considerable proportion of dark minerals should be called 'diorite'). Such writers define diorite as leucocratic (or felsic) and gabbro as mesotype to melanocratic (mafelsic or mafic), although they do not agree on the quantitative definition of these terms. Shand's boundary (1927, pp. 132, 184; 1939, pp. 196, 281) is 30 % by

volume, Lacroix's (1933, pp. 24, 192–3) 35 % by weight, Washington's (1923, p. 569)—for andesite and basalt—37·5 % by weight. E. Wenk (1946) has also recently gone on record in favour of a colour-index boundary between andesite and basalt, but without specifying an exact figure. Shand also uses the felspar criterion to distinguish between soda- and lime-diorites and soda- and lime-gabbros, while Johannsen also uses a colour-index boundary of 50 to mark off meladiorites and melagabbros. Later, Johannsen (1939, preface) has stated that he has considered substituting colour-index divisions of 10–40–70 for those of 5–50–95 used in his classification, although he gives no reason for this. Statistical evidence will be given in its favour later in this paper.

In addition to modal criteria, there are others based on chemical analysis; thus for glassy and merocrystalline rocks Lacroix uses the normative colour-index, defining andesites as belonging to class II and basalts to class III of the C.I.P.W. (Cross, Iddings, Pirsson, and Washington) system. Hatch and Wells, adapting the less rigid scheme of Brögger (1895), use a silica percentage of 55 (1937) or 52 (1926). Such definitions are of little use petrographically since they necessitate either a chemical analysis or an accurate mineralogical analysis combined with a chemical analysis of the constituent minerals, and this is impracticable except for a relatively small number of rocks.

For dealing with large series of rock specimens, types must if possible be defined in such a way that rocks can be named readily with a recourse to no more than thin-section or immersion micro-technique, yet so that chemically as well as mineralogically similar rocks are on the whole grouped together. In the framing of such a definition, boundaries should be selected so as to coincide as far as possible with minima in the frequency-distribution of rocks with relation to the chosen criteria. If such minima exist there will also be maxima which will correspond to real objective types. The investigation here recorded had as its object the establishment of such boundaries and types by the study of the frequency-distribution of actual rocks along co-ordinates representing the three main characters used in defining diorites and gabbros. Where no such boundaries can be shown to exist and it is still desired to divide a series into two or more types, the boundary selected may coincide with some chemical relationship or be a matter of practical convenience; if possible, both.

All the definitions of diorite and gabbro which have been quoted are claimed by their authors to include the more typical examples. Combining them, it appears that typical diorites are rocks in which oligoclase

or andesine is dominant and hornblende with or without biotite is subordinate in amount, while typical gabbros are rocks with labradorite or bytownite and a pyroxene in roughly equal proportions. If the anorthosites are also included, as rocks composed entirely or almost entirely of a lime-soda-plagioclase, we have the following tabular arrangement of types:

TABLE I.

Ferromagnesian minerals.	More sodic felspars.	More calcic felspars.
About equal to quartz and felspar (mafelsic) or predominant (mafic)	Mafelsic and mafic diorites or soda-gabbros	*Typical gabbros* and norites (mainly pyroxene-rocks), with some hornblende-rocks (hornblende-gabbros)
Definitely subordinate to quartz and felspar, but nevertheless essential (felsic)	*Typical diorites* (mainly hornblende and/or biotite rocks, with some pyroxene rocks (augite-diorites, &c.)	Felsic gabbros or lime diorites
Absent, accessory, or very minor in amount (holofelsic)	Holofelsic diorites or soda-anorthosites	*Anorthosites*

(The terms felsic, mafelsic, and mafic are here used qualitatively, as by Tyrrell in his 'Principles of petrology' (1926). They seem more satisfactory than the terms leucocratic, mesotype, and melanocratic, which have been given several quantitative meanings.)

The problem is that of deciding the appropriate quantitative values of the boundaries in this table, and also of correctly placing the mafelsic and mafic diorites and felsic gabbros. To do this, an attempt has been made to collect a random sample of rock modes within the limits of the field, excluding over-saturated rocks (limit, 10 % of quartz) and under-saturated (limit, $2\frac{1}{2}$ % of olivine and any felspathoid), and including only rocks with at least $\frac{2}{3}$ of the felspar as plagioclase (ranging in composition from An_{20} to An_{80}) and with up to 80 % of mafic minerals. Modes within these limits have been collected from all available literature. Gravimetric modes have been converted to volumetric, because the latter determine the visible character of rocks. Many authors publish modes without stating whether they are by weight or volume, and it has been necessary to decide this in many cases by indirect evidence. Such authors appear to think that the difference between percentages by weight and volume is insignificant; this may be true of rocks near the ends of the colour-index range, but for those with between 25 % and

75 % of dark minerals the difference may be anything from 4 % to 8 %, or even more if much iron ore is present.

The number of published modes being considered inadequate, as many rocks as possible in the British Museum collection, falling within the chosen limits, have been micrometrically analysed by means of the Leitz integrating stage, the composition of the felspar being determined by extinction methods, as these are likely to continue in use for ordinary determinative work on account of their speed and convenience. The average composition of zoned felspars was arrived at by estimating the composition and relative volume of the kernel and successive concentric shells of typical crystals. It is safe to say that by adding this set of data to that obtained from the literature a roughly representative sample of the saturated plagioclase-rocks has been obtained, although the total number (about 400 plutonic rocks) is, even so, smaller than is desirable. Similar (but scantier) data have been amassed for the hypabyssal and volcanic rocks, which will be considered after the diorites and gabbros.

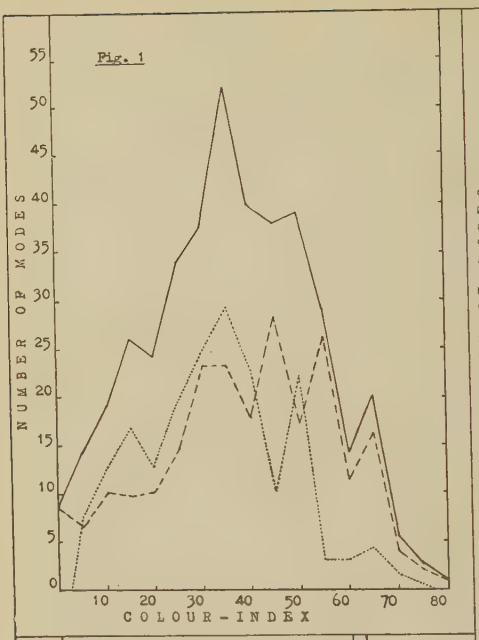
The data have been ranged in three series of subgroups according to the values of the three principal modal characters used as criteria, namely, (1) colour-index (volumetric percentage of dark minerals), (2) percentage of anorthite in the plagioclase, and (3) ratio of pyroxene to hornblende and biotite.

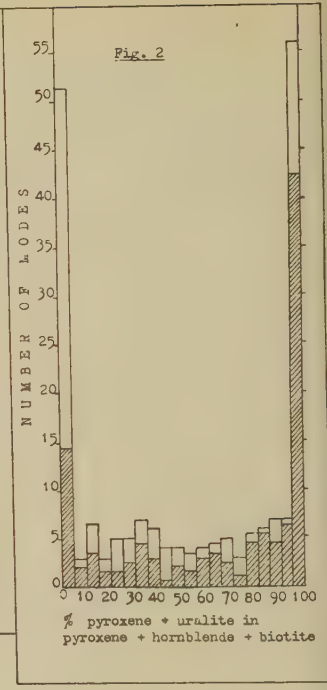
Fig. 1 shows the 418 modes in which the colour-index is known, approximated to the nearest 5 % and arranged as a frequency diagram. The distribution is also shown separately for rocks with oligoclase or andesine (Ab $>$ An), and with labradorite or bytownite (An $\geqslant$ Ab). The main diagram shows a fairly symmetrical distribution with a maximum at 35 and a range of 0–70, above which there are very few modes. The rocks with oligoclase and andesine also show a maximum value at 35, but are deeply divided at a value of 45 between this maximum and a secondary peak at 50, above which there is a very rapid falling off. Those with labradorite and bytownite, the bulk of which range from 30 to 65 (i.e. 28-67) are divided less deeply, but in three places. That at 40 appears to be the most significant of these, since it corresponds to a similar point in the labradorite- and bytownite-bearing dike rocks and lavas (figs. 11, 13); it divides the series into two nearly equal portions. This suggests the possible existence of felsic and mafelsic natural groups, the boundary between which is at a slightly higher colour-index for rocks richer in soda-felspar.

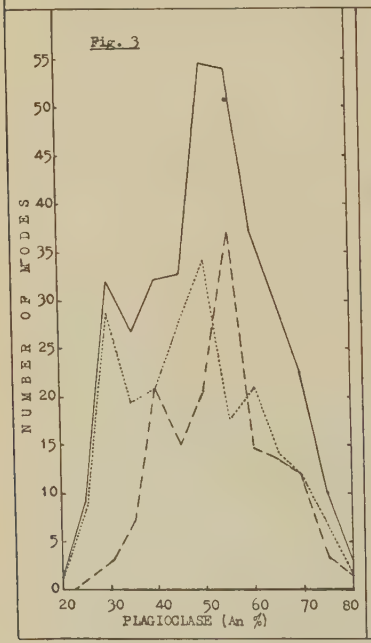
Fig. 2 shows the 362 modes in which the composition of the plagioclase is known (in 66 cases it is given as simply andesine, labradorite, &c.),

similarly arranged, rocks with colour-index values of less and more than 40 being also shown separately. No fewer than 55 rocks (about one-seventh) have plagioclase approximating to An_{50} in composition, this being actually the commonest value. This is partly because modes in which the felspar is described as andesine-labradorite have all been given this value (a similar explanation certainly accounts for the peak at oligoclase-andesine, An_{30}) but it is too marked a feature to be mainly explained away in this manner. This is true also for the felsic series considered separately: in this case there is a rapid decrease in numbers between An_{50} and An_{55}. The mafelsic series (colour-index > 40) has a maximum at An_{55} and falls away steeply and fairly symmetrically on both sides of this; there is a minor peak at An_{40}. This suggests that the An_{50} line is not a satisfactory boundary between major types, although one drawn between An_{50} and An_{45} might be so, especially for mafelsic types. It may be noted that a molecular proportion of 48 % of anorthite is equivalent to 50 % by weight; also that Holmes (1917) suggested that An_{47} would be a more convenient boundary than An_{50}, since it falls near the boundary between orders 3 and 4 in the C.I.P.W. system.

Fig. 3 shows all the data grouped at 10 % intervals according to the percentage of pyroxene + uralitic amphibole in pyroxene + hornblende + biotite. This indicates a marked division between the rocks with dominant pyroxene (i.e. those with 'dry' ferromagnesian minerals low in alumina) and those characterized by hornblende and biotite. About 57 % of the 415 rocks for which this ratio is given have values of 0–5 % or 95–100 % (i.e. in a clear majority of these rocks pyroxene is either exclusive of hornblende and biotite, or vice versa) and in less than 4 % of the total does the value lie between 45 % and 55 % (i.e. pyroxene about equal to hornblende and/or biotite). These values are about the same in subdivisions of the total according either to felspar or to colour-index; for rocks with andesine-labradorite (An_{50}) the proportion in which pyroxene is about equal to hornblende and/or biotite is still only 6 %. Biotite and hornblende are more commonly exclusive among soda-felspar rocks, pyroxene among lime-felspar rocks. (The boundary between Shand's metaluminous and subaluminous reaction types falls at or near 100 %). The nature of the dark minerals does, therefore, provide a basis for differentiating between two series of rocks, since in 96 % of the total number either pyroxene or hornblende + biotite is clearly dominant or exclusive. The two series, however, are not chemically distinct but parallel. The relatively sharp distinction between them suggests that something may be learned by studying their frequency-







Figs. 1 to 3.

DIORITES, GABBROS, and ANORTHOSITES.

Fig. 1 COLOUR-INDEX (Dark minerals, volume %)

..... Ab > An } Plagioclase
– – – – An > Ab }
——— Total

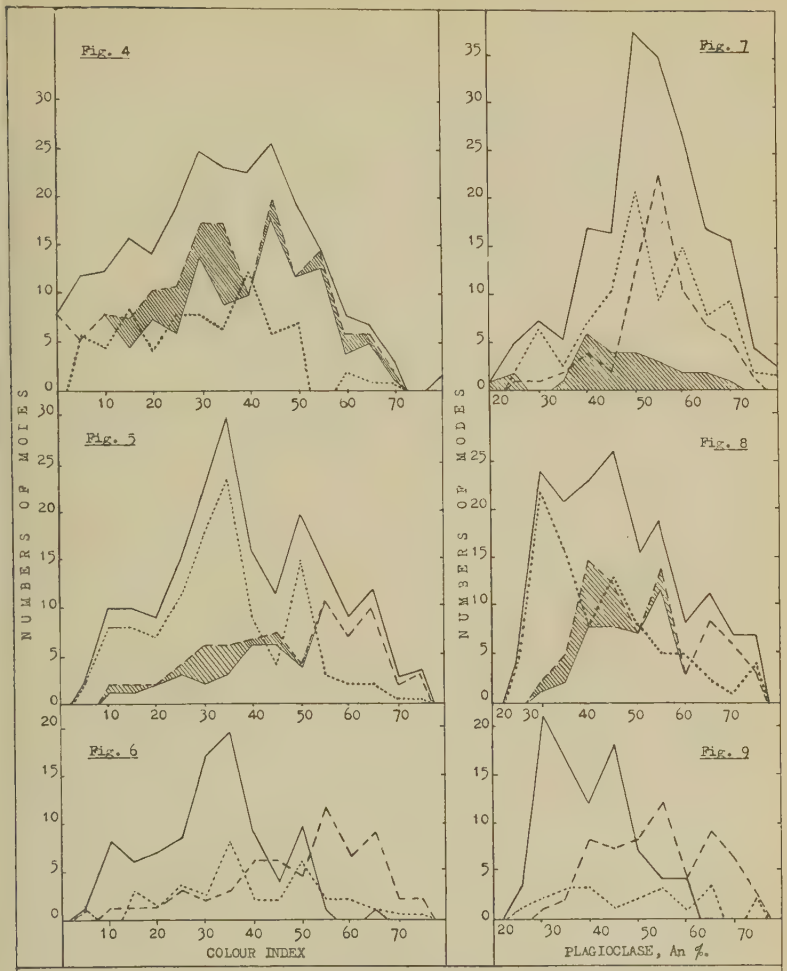
Fig. 2 COMPOSITION OF PLAGIOCLASE (An %)

..... Colour-index < 40
– – – – Colour-index > 40
——— Total

Fig. 3 NATURE OF DARK MINERALS, expressed as

$$\frac{\text{Pyroxene + uralitic amphibole}}{\text{Pyroxene + amphibole + biotite}} \times 100$$

Ab > An } Total
An > Ab }



Figs. 4 to 9. DIORITES, GABBROS and ANORTHOSITES.

	COLOUR-INDEX	PLAGIOCLASE, An %.
Pyroxene > Hornblende + Biotite	Fig. 4. ..... Ab > An (75 modes) _ _ _ An > Ab (159 modes) _____ Total (234 modes) Shaded An > Ab and Quartz > 2%.	Fig. 7. Colour-Index: < 10 (27 modes) Shaded; 10-40 (96 modes) .....; > 40 (73 modes) _ _ _ Total (192 modes) _____
Hornblende + Biotite > Pyroxene	Fig. 5. ..... Ab > An (111 modes) _ _ _ An > Ab (73 modes) _____ Total (184 modes) Shaded An > Ab and Biotite > 20% of dark minerals	Fig. 8. Colour-Index: < 40 (85 modes) .....; > 40 (71 modes) _ _ _ Total . . . (156 modes) _____ Colour-index > 40 and Biotite > 20% of dark minerals Shaded
	Fig. 6. Biotite, % of dark minerals: < 20 Ab > An .....; An > Ab _ _ _ > 20 _____	Fig. 9. Biotite, % of dark minerals: < 20 Colour-Index < 40 .....; > 40 _ _ _ > 20 . . . . . _____

distribution separately. This is done in figs. 4, 5, and 6 for colour-index and in figs. 7, 8, and 9 for felspar.

The colour-index diagrams all show the division at 40 (or 45) and also that this division is due to the more typical rocks—i.e. to those with labradorite and bytownite in the pyroxene series (fig. 4, dashes) and to those with oligoclase and andesine in the hornblende and biotite series (fig. 5, dots). If the quartz-bearing pyroxene-labradorite (and bytownite) rocks are excluded (shaded portion, fig. 4) the division is nearer 35 than 40 and the mafelsic types are clearly dominant. Among the hornblende- and biotite-andesine (and oligoclase) rocks, although the division is at 45, 40 is also a very low value (fig. 5, dots). A low frequency at a colour-index of 40 is, therefore, common to the typical members of both series. The division does not appear among the pyroxene rocks with oligoclase or andesine or among the hornblende and biotite rocks with labradorite or bytownite, but the former group is mainly felsic (fig. 4, dots) and the latter mainly mafelsic (fig. 5, dashes), and they overlap at a colour-index of 30–50; rocks of the hornblende-biotite series with labradorite or bytownite mainly have essential biotite if the colour-index is below 40, but are almost all without it if it exceeds 40 (dotted line and shaded portion, fig. 5).

Fig. 6 shows the strong correlation between essential biotite, soda-felspar, and a felsic character on the one hand and between absence of essential biotite, lime-felspar, and mafelsic character on the other; further, that among the rocks without essential biotite those with oligoclase or andesine are more numerous below a colour-index of 40, and those with labradorite and bytownite at and above a colour-index of 40.

This type of frequency-distribution suggests the existence of two overlapping groups, each of which if it could be considered separately would show a fairly symmetrical frequency-distribution about a typical value (in this case a colour-index of about 30–35 and 50–55 respectively). The position of the low value between the two maxima depends on the relative abundance of the two groups. Where the felsic group is dominant, as in hornblende and biotite rocks with dominant soda-felspar (fig. 5, dots) or those with essential biotite (fig. 6, continuous line), it lies at 45; where the mafelsic group is more abundant, as in pyroxene-labradorite rocks *without* essential quartz (fig. 4), it lies at 35. (The rather more marked displacement of the division among rocks of the hornblende and biotite series may also be due partly to the fact that these dark minerals contain the equivalent of pyroxene plus some felspar or felspathoid.) When one group greatly outnumbers the other the

former overlaps and conceals the latter (continuous line, fig. 1, in which the mafelsic rocks are swamped by the felsic). The colour-index value of 40 seems to mark a rough natural division, because it is the low value produced by the overlapping of the two groups when they are about equal in numbers (as in the case of the typical gabbros and the felsic gabbros *including* quartz-bearing varieties (fig. 1, dashes). It has the additional practical advantage that it is the point above which, in general, the dark minerals appear about equal in amount to the light ones.

Figs. 7, 8, and 9 show that the series with dominant pyroxene and with dominant hornblende or biotite overlap considerably as regards the composition of the felspar, most of the former lying between An_{40} and An_{70} and most of the latter between An_{30} and An_{60}. Many rocks of the pyroxene series have a value of An_{50} and the majority, whether felsic or mafelsic, lie above this value (fig. 7); An_{50} is actually the typical value for the felsic series and An_{55} for the mafelsic. Among rocks of the hornblende and biotite series the vast majority of those with essential biotite lie below An_{50} (fig. 9, and shaded portion of fig. 8), while most of those without essential biotite are mafelsic and have about 50 % or more of anorthite in the felspar, although many have less than this (fig. 9, dashes). Excepting for holofelsic rocks, which are almost confined to the pyroxene series, the point above which rocks in which the dominant dark mineral is pyroxene predominate over those in which it is hornblende or biotite lies between An_{50} and An_{45} (rather nearer the latter value than the former), and not at An_{50} as assumed by Rosenbusch when he substituted this limit between diorite and gabbro for one based on the dark minerals. The striking nature of the transition is brought out in fig. 10. At An_{45} there are twice as many rocks with dominant hornblende and/or biotite as with dominant pyroxene; at An_{50} there are only half as many.

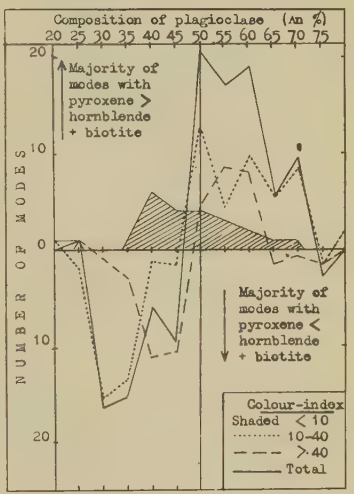


Fig. 10

Figs. 4–9 therefore constitute a case for utilizing a colour-index of about 40 and a plagioclase composition of An_{47-48} as the limits in table I.

These limits should not be too rigidly applied, especially as neither the colour-index nor the composition of the felspar can be very accurately determined. In applying such limits it would seem best to place rocks falling on or very near them with the adjacent types with which their affinity is greatest. For example, the more typical diorites (the mica-diorites of Rosenbusch) may be permitted to extend up to An_{50}. Rocks falling near the colour-index boundary of 40 should be grouped with the diorites if their felspar is more sodic and with the gabbros if it is more calcic; thus diorites will in practice range up to rather more than 40 and gabbros down to rather less than 40.

It is suggested that the term *meladiorite* should be used for mafelsic diorites or soda-gabbros; this only slightly extends the sense of the term as used by Johannsen and by Niggli, whose lower limit for it is a volumetric colour-index of 50, whereas the limit of 40–45 here proposed corresponds roughly to 50 % *by weight* of dark minerals. Similarly the felsic gabbros may be termed *leucogabbros*; this term is used by Niggli in practically the same sense. The prefix *leuco*, implying a colour-index lower than usual, is normally applied to holofelsic rocks, because most igneous rock-types are felsic, but it seems quite logical to use it to indicate the felsic variants of mafelsic types.

There appears to be no definite discontinuity between the holofelsic and felsic rocks (fig. 4, dotted line), but practically all the rocks in the data called anorthosite contain some dark minerals, ranging up to 10 % or rather more. As this is the conventional limit used in most classifications it will be adopted here as the boundary between holofelsic and felsic types. The distribution of the holofelsic rocks, so defined, with relation to the composition of the felspar (shaded portion of fig. 7) shows a discontinuity between the oligoclasites on the one hand and the andesine- and labradorite-rocks on the other, but no division near An_{50}. It appears from this diagram that andesine-rocks are more common than labradorite-rocks, but this is due to the overweighting of the data with Buddington's modes (1944) from the predominantly sodic Adirondack anorthosite, and to the fact that many published descriptions of anorthosites quote the felspar as labradorite without giving its composition, and cannot therefore be used in fig. 7. If these are taken into consideration, it appears that labradorite-rocks predominate over andesine-rocks among published anorthosite modes, which therefore seem to have a range and distribution similar to that of rocks of the pyroxene series in general. Accordingly it is proposed that the An_{47-48} boundary be used in the same manner as for other rocks, the term anorthosite being used alone

to signify rocks composed predominantly of plagioclase ranging from andesine-labradorite to labradorite-bytownite, those composed of an andesine with well under 50 % of anorthite being called andesine-anorthosite or andesinite. The collective term *leucodiorite* (used by Johannsen in a narrower and by Niggli in a broader sense) may be used to cover oligoclasite and andesinite. Rocks falling near the colour-index limit of 10 should be placed with anorthosite rather than leucogabbro, since anorthosite is to be considered a major type on account of its quantitative importance.

As an upper limit to the mafelsic diorites and gabbros a colour-index of 70 is suggested because while there are many modes with an approximate colour-index of 65 there are very few indeed with one of 70 or more. Rocks with more than about 70 % of dark minerals should be called gabbro-pyroxenites, &c. Among the many modes of pyroxenites and hornblendites of different kinds listed by Tröger (1935), the vast majority have 100 % of dark minerals and are truly holomafic. We have, therefore, the colour-index ranges suggested but not adopted by Johannsen (1939), of 0–10–40–70–100.

A consideration of hypabyssal and volcanic rocks indicates that the same divisions can be used for them. For the purposes of this investigation, the grain-size range proposed for the gabbro-dolerite-basalt series by the British Association Committee on Petrographic Nomenclature (A. K. Wells, 1936) has been adopted. A collection of 115 modes of 'hypabyssal' rocks, so defined, has been made and they have been plotted in two series (those with dominant pyroxene and those with dominant hornblende or biotite respectively) in fig. 11 for colour-index and in fig. 12 for felspar. The rocks with dominant pyroxene, mainly dolerites (96 modes), far outnumber the others. They have a high concentration about a colour-index of 45; there are many below 40, but these are mostly 'granodolerites' containing much micropegmatite. Practically all dolerites have a plagioclase of composition between An_{48} and An_{67}. Data for microdiorites and diorite-porphyries are scarce, and it was found that most of the rocks so called in the British Museum collection were either microgranodiorites or granodiorite-porphyries, or devitrified andesites or dacites. Of the few of which the modes lie within the chosen limits, practically all have felspar with 40% or less of anorthite. They show no clear division as regards colour-index, but if their numbers are multiplied by five to make the total comparable with that of the dolerites, 40 is the value below which rocks with dominant hornblende or biotite and above which those with dominant pyroxene are predominant.

Adequate data for the volcanic rocks have been difficult to obtain because of the abundance of merocrystalline types in the dacite-andesite series, and of the relative scarcity of silica-saturated basalts. As it is no attempt has been made to exclude rocks which have, or may have more than 10 % of normative quartz occult in a glassy matrix, the only rocks excluded as dacites being those so defined by Rosenbusch (1907 p. 993) by the presence of first-generation quartz. There is accordingly a greater proportion of rocks of low colour-index in the data for volcani than for plutonic and hypabyssal rocks, despite the absence of volcani equivalents of the anorthosites. Few modes could be found for basalts because most basalts contain some olivine; and few authors have con sidered it worth while to attempt quantitative mineralogical analyse of andesites, which normally consist largely of very minute felspars i a glassy matrix. A few andesites were selected from the British Museur collection as suitable for micrometric analysis, and the proportions o phenocrysts and matrix, and of different constituents of the matrix were estimated separately. A rough correction was applied to the result obtained for pyroxene and 'ore'-grains in the matrix, to compensate fo the fact that these are normally less in diameter than the thickness of th section. In some cases it was necessary to estimate by eye the approxi mate composition of the matrix. The few fresh saturated basalt available were also measured.

Plotting the results (109 modes) together on the colour-inde co-ordinate, the frequency-distribution shown in fig. 13 was revealed giving once again a deep division at 40, with a peak at 10 due to th andesites and at 50 due to the basalts. The low typical value for andesite may be due not only to the presence of many rocks with occult quart but to the fact that the glassy matrix also contains occult dark mineral To obtain a frequency-distribution more directly comparable with tha of the intrusive rocks, an attempt was made to correct the colour-inde of merocrystalline rocks for this. By a comparison between the mode and norms of rocks for which both were available, it was found that o the average the potential colour-index of the glass is about 20 % of th actual colour-index of the crystalline material considered separately If the colour-index values are corrected in this way, the result is th frequency-distribution shown by the line of dots in fig. 13 with typical value ranging from 10 to 20 and a minor peak at 30. This line dots represents the hyalopilitic and pilotaxitic lavas, which roughl coincide with the andesites and which are distinguished texturall because most lavas with plagioclase have pyroxenes as their dominar

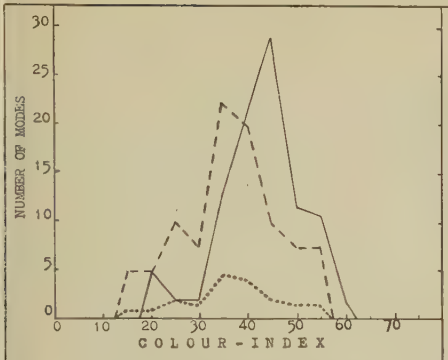


Fig. 11. DOLERITES and MICRODIORITES
——— Pyroxene > hornblende + biotite (97 modes, mainly dolerites).
..... Hornblende + biotite > pyroxene (19 modes, mainly microdiorites and lamprophyres).
– – – – The same (scale five times larger).

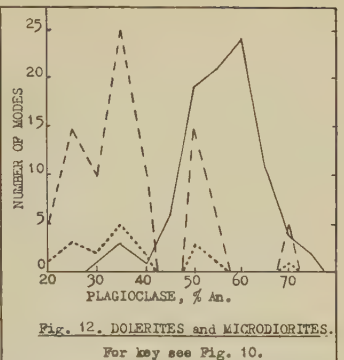


Fig. 12. DOLERITES and MICRODIORITES.
For key see Fig. 10.

Fig. 13. ANDESITES and BASALTS
——— Total, percentage of actual dark minerals.
..... Hyalopilitic and pilotaxitic } corrected for
– – – Intersertal and intergranular } occult dark minerals

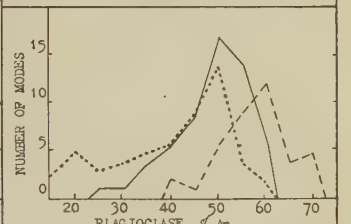
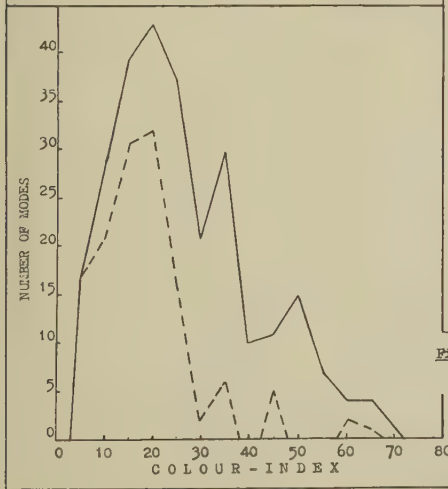


Fig. 14. ANDESITES (including cryptodacites)
..... Plagioclase of matrix.
– – – – Phenocrysts
——— Average plagioclase developed.



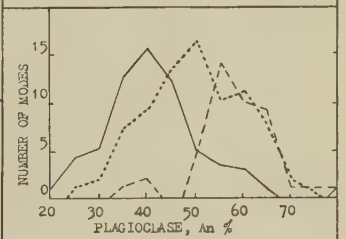


Fig. 15. ANDESITES and BASALTS
——— Estimated potential plagioclase in 63 hyalopilitic and pilotaxitic rocks.
..... Normative plagioclase in 335 rocks called basalt. Scale five times reduced. (After Bowen).
– – – Modal plagioclase in 46 intersertal and intergranular rocks ("basalts")

Fig. 16. THE GRANODIORITE-DIORITE SERIES.
——— 269 modes of hornblende and biotite-granodiorites, quartz-diorites and diorites (including mafelsic types).
– – – Granodiorites and quartz-diorites alone.

dark minerals; the other line (dashed) represents the intersertal and intergranular rocks, mainly basalts, which are almost unaffected by the correction. The result is still to show that there are few lavas with a colour-index of 40, and that below this andesitic types and above it basaltic types predominate. The few andesitic types with a higher colour-index than 40, and the basaltic types with a lower colour-index, are all characterized by labradorite.

It is difficult to compare the composition of the felspar in andesitic types with that in basalts, because of the great difficulty of estimating without analysis the average composition of the total (including occult) felspar of andesites, or even of the felspar actually developed. Fig. 14 (dashes) shows that the phenocrysts of andesites are generally of labradorite and are typically more calcic than the average (normative or modal) felspar of basalts (fig. 15, dashes). The matrix felspar is much more sodic but is nevertheless more calcic than the average felspar of diorites (fig. 8, dotted line). The felspar actually developed, obtained by averaging these two, virtually coincides with Bowen's curve (fig. 15, dotted line; Bowen, 1927, p. 140, fig. 40) showing the frequency-distribution of normative plagioclase in 335 rocks called basalt. An attempt was made to estimate the frequency-distribution of the average potential (including occult) plagioclase in 63 andesites. The residual glass was assumed to contain at least half of its volume of plagioclase of a composition somewhere between that of the matrix plagioclase and An_{10}, and maximum and minimum values between which the potential average plagioclase probably falls were found for each rock. The means of these respective maximum and minimum values were plotted as a frequency diagram, which is shown in fig. 15 in conjunction with that for the plagioclase actually developed in 46 rocks of basaltic type. The respective typical values are An_{40} for andesitic and An_{55} for basaltic types, the former being mainly below and the latter mainly above An_{50}, though they overlap at this point. The frequency-distribution of the potential plagioclase has the same range as that of the matrix plagioclase (fig. 14), but the maximum frequency of the latter is at a higher value, i.e. An_{50}. The symmetrical nature of the frequency diagram for the potential felspar of andesites suggests that the method of estimation used, while rough and ready, is accurate enough for practical purposes. If it indicates a predominance of albite over anorthite, the rock in question should be called andesite-basalt (e.g. alboranite, miharaite, mijakite). Some rocks of basaltic type (e.g. Markle basalt) have an abnormally low colour-index owing to enrichment in felspar phenocrysts, and should be called

leucobasalts. The mafelsic lavas with oligoclase and andesine are all of basaltic type and should be called oligoclase-basalts (e.g. spilites) and andesine-basalts.

The same limits, with the same proviso as regards overlap of boundaries, can therefore be applied to the lavas as to the intrusive rocks, using texture instead of the nature of the dark minerals as one of the diagnostic characters.

Chemical data.

About a third of the modes collected from the literature were accompanied by chemical analyses, which are also available for a few of the rocks of which the modes have been micrometrically determined. These analyses have been plotted in various ways, using distinctive symbols to indicate the dominant dark minerals, the composition of the felspar, and the relative abundance of the dark and light minerals, in order to show how far the limits suggested for various types reflect chemical distinctions.

Since the field under discussion only covers rocks with plagioclase as the predominant felspar and at or slightly above the silica-saturation level, it is not to be expected that any definite segregation of types can be demonstrated about any given silica percentage. A study of silica percentages reveals that while Hatch's limiting value of 52 % is an approximate upper limit to mafelsic rocks (using the colour-index of 40 as the boundary between felsic and mafelsic), there are many rocks with a lower silica content which most petrographers would agree to call diorites (i.e. rocks with dominant andesine and subordinate hornblende and biotite), while many typical quartz-gabbros have a higher silica content. There is a rather more definite segregation of the lavas, nearly all basalts and andesite-basalts having less than about 54 % SiO_2 and nearly all andesites having more than this. As the relevant diagrams show such a marked overlap of types they have not been reproduced.

The relation of the modal limits proposed to chemical composition is best demonstrated by means of the Niggli values[1] *al-fm-c-alk*, which express the general mutual relationships of the bases. This can be fully shown on two triangular diagrams each of which is a projection of the *al-fm-c-alk* tetrahedron. Figs. 17, 18, and 19 (plate XXV) show all the analyses plotted on the $al \,\lceil alk : fm : c$ triangle and figs. 20, 21, and 22 on the $c + fm : al : alk$ triangle. Both sets of triangles show that no single one of the characters which have been used to define diorites and gabbros

[1] $al : fm : c : alk = Al_2O_3 : 2Fe_2O_3 + (Fe,Mg,Mn)O : (Ca,Ba,Sr)O : (K,Na)_2O$ (molecular values); $al + fm + c + alk = 100$.

actually separates chemical types; the least satisfactory character from this point of view is the nature of the dark minerals, and the most satisfactory the colour-index. If, however, all the characters are taken into account, it can be shown that the modally defined types here suggested correspond quite closely with definite fields in the Niggli triangles, allowing for a certain amount of marginal overlap. The few exceptions are nearly all mineralogically unusual rocks, or rocks lying on or near the modal limits suggested; if therefore they had been given symbols according to the principle of placing rocks on or near a modal boundary with the adjacent major type which they most resemble, the correspondence would be even closer.

Thus in fig. 17 (the $al+alk:fm:c$ triangle for diorites, gabbros, and anorthosites) it can be seen that for all the holofelsic rocks $al+alk > 4fm$; for oligoclasites and andesinites $al+alk > 2c$; for typical (labradorite-) anorthosites and bytownite-anorthosites $al+alk < 2c$; andesinites and typical anorthosites lie close together on either side of the line $al+alk = 2c$, reflecting the fact that they constitute a natural group with the plagioclase averaging about An_{50}. For most diorites, $al+alk < 4fm, > fm$; exceptions with $al+alk < fm$ all have $al+alk > 2c$ (i.e. they are types with fm high at the expense of c). Gabbros and meladiorites all have $al+alk < fm, < 2c$. For leucogabbros $al+alk < 2c$, $> 0{\cdot}25fm, < 4fm$. For hornblende- and biotite-diorites $al+alk > 2c$. Thus hornblende- and biotite-diorites are almost quantitatively marked off from gabbros and meladiorites; leucogabbros overlap gabbros and pyroxene-diorites, while pyroxene-diorites overlap leucogabbros and hornblende- and biotite-diorites.

In fig. 18 the dolerites correspond in position to the gabbros, and a number of dioritic lamprophyres fall with them. Hypabyssal equivalents of the typical diorites are relatively uncommon. Among the volcanic rocks (fig. 19), the andesites nearly all have $al+alk > 2c$, $> fm$, $< 4fm$; the basalts have $al+alk < 2c$ and $< fm$; for andesite-basalts $al+alk < 2c, > 0{\cdot}25fm$ and $< 4fm$.

In fig. 20 (the $c+fm:al:alk$ triangle for diorites, gabbros, and anorthosites) all the analyses show $al > alk$ (this demonstrates the absence of peralkaline rocks from the field under consideration). The holofelsic rocks all have $al > 40$. For gabbros, meladiorites, bytownite-anorthosites, and some leucogabbros, $c+fm > 7alk$. For diorites, anorthosites, andesinites, oligoclasites, and the rest of the leucogabbros, $c+fm < 7alk$, $> alk$. For leucogabbros and anorthosites (labradorite-rocks) $al > 2{\cdot}5alk$; for hornblende- and biotite-diorites, andesinites, and oligoclasites

$al < 2{\cdot}5alk$. As in fig. 17, the andesine- and labradorite-anorthosites lie close together.

In fig. 21 the compact distribution of the dolerites reflects their remarkable uniformity of composition. They are fairly definitely marked off from the microdiorites and dioritic lamprophyres by the line $c+fm = 7alk$. Among the volcanic rocks (fig. 22) it seems that a better separation of the basalts from the andesites is given by the line $c+fm = 6alk$ or $5alk$ than by the line $c+fm = 7alk$, but for nearly all the andesite-basalts, as for the leucogabbros, $al > 2{\cdot}5alk$, while for andesites $al < 2{\cdot}5alk$.

The principal points which emerge from these diagrams are, therefore, that meladiorites are not generally chemically distinguishable from gabbros; that typical diorites with oligoclase or andesine and subordinate hornblende or biotite are quite distinct both from gabbros and leucogabbros; that some pyroxene-diorites and some leucogabbros are intermediate chemically between typical diorites and typical gabbros; and that holofelsic rocks form a series distinct from the rest and clearly divisible into different felspathic types along the line from $al+alk$ to c, andesinites and anorthosites lying close together and evidently forming a single natural group. The modal limits suggested seem justified on the basis of the distribution of chemical data; even the two analyses of rocks with a colour-index exceeding 70 are distinguished from all the others in fig. 17 by having $fm > 4(al+alk)$, this division corresponding to that of the holofelsic rocks at the opposite side of the triangle.

Interpretation and suggestions for nomenclature.

The distribution of the modal data, while providing a basis for the delimitation of major types, also calls for explanation. It is suggested that the twofold grouping with relation to colour-index is the same as that pointed out by Daly (1933, p. 39) among calc-alkalic rocks in general: that of granite-granodiorite and andesite on the one hand, and gabbro, dolerite, and basalt on the other. The bulk of *typical diorites* are really the basic end of the granite-granodiorite series; their typical occurrence is as marginal facies or cupolas of granodiorite masses or as small stocks or bosses—i.e. as 'subjacent' rather than 'injected' masses (Daly, 1914, pp. 382–383; 1933, pp. 454–455). Average diorite is midway in chemical composition between average basalt and average granite (Daly, 1914, p. 382). Diorites, therefore, in general have a composition consistent with derivation from two parent materials, one of granitic and the other of basaltic composition, in approximately equal proportions; the result,

perhaps, of the assimilation of sialic granite by simatic basalt magma followed by gravitational differentiation, as according to Daly (1914, 1933), or perhaps of the granitization of pre-existing basic rocks. The typical gabbros, dolerites, and basalts, on the other hand, seem to be truly the result of the solidification of a primary magma; the differences which they show from the oceanic olivine-basalts, which are presumed to be the typical simatic rocks, are such as can be explained by gravitational differentiation or by a comparatively slight contamination with sialic material such as one would expect to result from their characteristic mode of occurrence (dikes, sills, and laccoliths, i.e. floored intrusions presumed to have cooled relatively quickly). In the case of some norites, the contamination has been by aluminous sediment (Read, 1923, 1924) while hornblende-gabbros probably result from the ingestion of material rich in volatiles (Daly, 1933, p. 409). On this hypothesis of two radically different modes of origin, one would expect that while all sorts of gradations between typical diorite and typical gabbro should actually occur, they should be quantitatively less important than either type, as is in fact the case.

Since diorites are typically characterized by essential hornblende and biotite (the 'mica-diorites' of Rosenbusch) those with hornblende alone should always be called 'hornblende-diorite'. The *augite-diorites* are often associated with anorthosites, and tend to fall near them in the chemical diagrams. The 'gabbroic anorthosites', which are so called because they form marginal facies of anorthosite masses and which some authors (e.g. Harrison, 1944; Vogt, 1924) consider to represent their parent magma, are in many cases augite-diorites and may be called anorthosite-diorites. Augite-diorites pass with increasing anorthite content of the felspar into Spurr's 'belugites'. The type of this name quoted by Tröger (1935, p. 146) is simply a felsic gabbro with andesine-labradorite. The corresponding lava, *aleutite*, is not to be modally distinguished from pyroxene-andesites generally.

The meladiorites are chemically gabbroic, excepting when rich in alkali-felspar, biotite, or quartz. If chemically gabbroic, they should be given names which indicate their closer affinity with the gabbros than with the diorites. Thus the few rocks with dominant pyroxene in this division practically all have a basic andesine and are best called andesine-gabbros (or, if hypersthene is predominant, andesine-norites). The hornblende-meladiorites, which form the majority, are not distinctly marked off from the hornblende-gabbros; they form with them one series with felspar ranging mainly from An_{40} to An_{65} and with a typical

value of An_{50-55}. There seems no reason why they should not be grouped under the general term *bojite*. This name was coined by Weinschenk (1898) to denote gabbros with dominant primary hornblende, and is so used by Johannsen (1937, p. 226); but Tröger (1934) discovered that the felspar of Weinschenk's type rock (from Pfaffenreuth, Bavaria) is medium andesine and therefore redefined bojite as 'dark hornblende-diorite'. It has been used recently in important papers by Pulfrey (1946) in the first sense and by Barth (1945) in the second, and has thus established itself in petrological literature with two mutually exclusive meanings. The obvious solution is to recognize that there is no essential difference between the gabbroic hornblende-meladiorites (i.e. those which do not contain essential quartz, biotite, or alkali-felspar) and the hornblende-gabbros, and to use the term bojite for both. If desired, those with andesine can be distinguished as andesine-bojites, this being nearly synonymous with hornblende-meladiorite. So used, bojite is synonymous with 'diorite' as originally defined by von Leonhard (1823, p. 104) and as used megascopically by the authors of the Quantitative Classification (1903, p. 183)—a sense which necessarily excludes typical diorite as currently used. The term bojite should not be used for meladiorites with abundant biotite or for those of appinitic type, since these are too alkali-rich to be chemically gabbroid.

The leucogabbros, though numerically important among the data, have not been treated as a major type because their concentration at a colour-index of 30–35 is probably due to the overlap of minor variations of the adjacent major types. Firstly, there are rocks resembling typical mica-diorites (but usually pyroxene-bearing), excepting that their felspar is labradorite. Such rocks, while included under the general heading 'leucogabbro', may be called labradorite-diorites. Secondly come the numerous quartz-gabbros and granophyric gabbros, which have been included in the data because they mostly contain less than 10 % of quartz. Both of these groups have a colour-index distribution like that of the typical diorites, with a maximum frequency of about 35. Thirdly come the basic marginal facies of anorthosite masses, ranging in colour-index up to about 30 but rarely above this; these may be called anorthosite-gabbros, anorthosite-norites, or anorthosite-bojites, according to the nature of their dark minerals. Lastly come those leucogabbros which are typical in the sense that they are felsic variants of normal gabbros; they include many norites (leuconorites) the felsic character of which reflects their origin as a result of the contamination of gabbroic magma by aluminous sediments.

The few dolerites with a colour-index of about 35 or less are all granophyric types with considerable micropegmatite and a felspar ranging down to andesine. Such types should be given the distinctive name *granodolerite* (Shand, 1917; Holmes, 1920). They have a range in colour-index close to that of the quartz-gabbros. Dolerites with a plagioclase more sodic than andesine-labradorite should be distinguished as andesine-dolerites or oligoclase-dolerites. The hypabyssal hornblende and biotite-rocks include a few lamprophyres (kersantite, spessartite, &c.) which are mafelsic types, but some of which overlap the colour-index boundary of 40. Here again the limits should not be applied too rigidly, especially to types like dolerites and lamprophyres, which are more clearly definable on a textural and qualitative than on a quantitative basis. Nevertheless, some rocks have been called kersantite or spessartite which, on account of their felsic character, should have been called 'microdiorite'. *Malchite* has been described variously and inconsistently as diorite-aplite and as a lamprophyre (Hatch and Wells, 1937, p. 251; Johannsen, 1937, vol. 2, p. 402), but Tröger's type specimen (1935, p. 147) occupies exactly the same position on the chemical diagrams as his typical diorite and andesite; it is, in fact, a typical microdiorite.

The nomenclature and delimitation of the volcanic types has already been discussed, excepting for one point: the separation of over-saturated types in which the quartz is occult in the residual glass. The majority of rocks generally described as andesites probably fall into this category. It is obvious from a comparison between fig. 13 and figs. 4 and 5, as well as between the chemical diagrams for plutonic and volcanic rocks, that most andesites, as defined by texture and by the minerals actually developed, are more felsic than most diorites. As their residual glass, as proved by analyses, commonly has the composition of a mixture of quartz and alkali-felspar, it may be said in general that the bulk of andesites correspond with granodiorites rather than diorites, a conclusion reached by Tyrrell (1921) from a consideration of chemical analyses. If, therefore, the term dacite is used in Rosenbusch's sense to mean andesites which are siliceous enough to be characterized by first-generation quartz, the dacite-andesite series as a whole can be considered to correspond to the granodiorite-diorite series, the boundary between dacite and andesite corresponding to a higher silica content than that between granodiorite and diorite. A close correlation can be demonstrated between the two series by combining the modal data for hornblende- and biotite-diorites, and meladiorites (fig. 5) with the data given for granodiorites and quartz-diorites by Johannsen (1937, vol. 2),

Shand (1939), and Tröger (1935), and plotting the combined data as a colour-index frequency diagram (fig. 16). It will be seen that the frequency-distribution revealed by this diagram corresponds very closely with that shown for andesites and basalts in fig. 13. The maximum at 15–25 is rather higher than the corresponding value for andesites; this is to be expected, because hornblende and biotite correspond to pyroxene and iron ore plus some alkali constituents, which in typical andesites are present in the glassy matrix. The peak due to meladiorites corresponds to that of the basalts; it is relatively less important because the meladiorites constitute only a relatively small and aberrant part of the gabbroic rocks. The minor peak at a colour-index of 30, present in both diagrams, is probably due to overweighting with diorites and andesites of dioritic composition, since no granodiorites additional to those found in the literature mentioned are included, and dacites with first-generation quartz are excluded. This parallelism between the frequency-distribution of the plutonic and volcanic rocks tends to bear out the generally accepted genetic connexion as well as the chemical parallelism between the granodiorite-diorite series and the dacite-andesite series.

While admitting the difficulty of exactly delimiting the over-saturated andesite lavas with occult quartz, it should be possible to make a rough distinction by consideration of the refractive index and relative abundance of the glass. More data are required fully to work out the possibilities of this, but we know from the work of Tilley (1922, p. 27) that typical rhyolite-obsidian has about 25–30 % of normative quartz and a refractive index of less than 1·500, while the refractive index of trachyte-obsidian ranges from 1·505 upwards. If, therefore, the residual glass has a refractive index of less than 1·500 and amounts to more than one-third of the volume of the rock, the latter almost certainly contains more than 10 % of potential quartz. Such rocks should be called cryptodacites (= dacitoid, Lacroix, 1933, pp. 33, 192) to distinguish them from the true dacites with crystalline quartz. It is probable that most rocks of andesitic type with an abundant glassy matrix are really cryptodacites.

The majority of the coarse-grained rocks falling within the limits adopted in this paper contain little or no orthoclase. Those with essential orthoclase (the conventional lower limit of which is 13·3 % of the total felspar) should be distinguished as monzonitic diorites or gabbros (monzodiorite, monzogabbro). Most of them have pyroxene, generally with biotite, as the principal dark mineral; such monzonitic pyroxene-diorites have been given the name mangerite (Kolderup, 1903). The terms syenodiorite and syenogabbro are ambiguous because they are

often used (e.g. by Hatch and Wells, 1937, pp. 196, 233) as synonymous with monzonite. Among the volcanic rocks, the andesites typically contain considerable occult orthoclase, which with the hypersthene and iron ore is equivalent to the quartz and biotite of the quartz-mica-diorites ('tonalites'); it is, therefore, unnecessary, and in practice impossible, to distinguish a group of orthoclase-bearing andesites, although holocrystalline types in which much orthoclase can be seen in the groundmass may be called trachytic andesites. Among the basalts, most of the orthoclase-bearing types also contain olivine and felspathoids, and are, therefore, outside the scope of this paper.

The boundaries suggested for the major types considered in this paper may seem indefinite and irregular to those accustomed to rigid and symmetrical pigeon-hole classifications, and who, perhaps, deny the existence of objective rock types. These critics should re-read the last chapter of Bowen's 'Evolution of the igneous rocks', in which he defends the traditional modal, loose classification. It is claimed that the present paper demonstrates within its limited field the existence of real objective types, which are in general those of the traditional classification; and that it roughly fixes rational quantitative limits to them which can be easily ascertained modally. The limits suggested have the advantage that, apart from distinction of meladiorite and leucogabbro from typical diorite or gabbro, they involve very little alteration to the names used for existing rocks. The system which comes nearest to the proposals made here is that of Niggli; indeed, if the colour-index limits of the Niggli system were weight instead of volume percentages they would be nearly equivalent to the volume percentages here used. The colour-index limits of 10–40–70 could, of course, be quite easily applied within the framework of the Shand or Johannsen systems.

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PROCEEDINGS

OF THE

MINERALOGICAL SOCIETY

(1948)

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., January 22, 1948, *at* 3.15 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

THE following were provisionally elected Ordinary Members of the Society: John Graham Comrie Anderson, John Angove, William Anderson Archibald, Norman Owen Clark, Norman Mederson Comber, Duncan Leitch, Ronald Keith Mitchell, William Anderson Mitchell, Joseph Patrick Roberts, Arthur William Scoble, Arnold Nigel Smith.

The deaths of Lord Rayleigh and Dr. W. R. Schoeller were announced. The names of R. B. Fisher and G. M. Yatsevitch were removed from the List of Members.

Reports were received from the Treasurer and the Editor. The Treasurer presented the audited accounts for 1947, which were approved.

Proposals for the modification of Bye-laws 5, 16, 17, and 18 were considered.

Dr. F. A. Bannister was appointed to represent the Society on the National Committee of the International Union of Crystallography.

The desirability of holding a Special Meeting of the Society during the International Geological Congress was discussed.

SPECIAL GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., January 22, 1948, *at* 5 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

The following alteration to the existing Bye-law 3 was approved:

Bye-law 3. Every candidate for Ordinary Membership shall be proposed by two Members, one, at least, of whom shall have personal knowledge of the candidate.

The President indicated that the Council had considered the new Bye-law 5 at its meeting earlier in the afternoon and had thought it necessary to propose the following additional words so as to preclude the possibility of a member compounding when in arrears with his annual subscription. He therefore moved from the Chair the following amendment. That the words 'provided always that no member shall be entitled to compound who is in arrear with his annual subscription for the preceding calendar year' be added at the end of the proposed new Bye-law 5, as already circulated to members. The amendment was carried *nem. con.* and the following Bye-law was approved:

Bye-law 5. Ordinary Members shall pay an entrance fee of one guinea and an annual subscription of one guinea. They shall be entitled to compound for the annual subscription by a single payment of £30, but members who have paid their annual subscriptions for fifteen years shall be entitled to compound for £20 and those who have paid such subscriptions for twenty-five years shall be entitled to compound for £10, provided always that no member shall be entitled to compound who is in arrear with his annual subscription for the preceding calendar year.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., January 22, 1948, *at* 5.15 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

Fifty-two Members and nine Visitors were present.

The elections of eleven Ordinary Members of the Society provisionally made by the Council were confirmed.

The following exhibits were shown:

1. Colour changes in gemstones irradiated by X-rays: by Mr. R. Webster.

2. Silica-glass evaporating dish coloured by radioactive salts: by Mr. P. A. Sabine.

The following papers were read and discussed:

1. A note on the thermal decomposition of chrysotile: by Dr. M. H. Hey and Dr. F. A. Bannister (vol. 28, p. 333).

2. Dickite and collophane in the Magnesian Limestone of Durham: by Dr. K. C. Dunham, Dr. G. F. Claringbull, and Dr. F. A. Bannister (vol. 28, p. 338).

3. Sillenite from South Africa: by Dr. F. A. Bannister.

4. A simple dichroscope: by Dr. A. T. J. Dollar.

CLAY MINERALS GROUP

Held in the Apartments of the Geological Society, Burlington House, London, W., January 23, 1948, 9.55 *a.m.* and 2 *p.m.*

Dr. G. W. Brindley in the Chair.

About seventy Members and Visitors were present. The meeting was devoted mainly to a discussion of 'Clay minerals as catalysts', followed, in the afternoon session, by general papers relating to industrial adsorption. The second Annual General Meeting on business matters of the Group was held at 5 p.m.

The following papers were read and discussed (abstracts in Clay Minerals Bulletin, no. 2, August 1948):

1. Clay minerals as catalysts: general introduction: by R. H. S. Robertson.
2. The use of activated clays as catalysts in polymerization processes: by F. E. Salt.
3. The use of montmorillonite catalysts in the cracking of petroleum fractions: by J. N. Haresnape.
4. The use of clays for fluid catalytic cracking (with film and demonstration): by T. H. Blakely.
5. The adsorption of dyestuffs by montmorillonites: by Miss B. S. Emödi.
6. The use of clays as bleaching agents, with a note on catalytic usage: by A. Ibison.
7. Separation of long-chain and compact molecules by adsorption on attapulgite-containing clays: by J. Nederbragt.
8. Vapour-pressure/capillarity/temperature relationships in clays: by B. Vassiliou and J. White.
9. Some notes on free energy relations in clay complexes: by D. M. C. MacEwan.

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., April 1, 1948, *at* 3.15 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

The following were provisionally elected Ordinary Members of the Society: Frederick William Bird, John Frederick Thomas Blott, George Charles Harry Clark, David Vanner Crawford, Rex William Grimshaw, John Raymund Foggan Joyce, Robert Joseph King, Edgar Wyndham Powell, William Rawlinson, Henry Martin Richardson, Robert Kenworthy Schofield, and Helen Towers.

Reports were received from the Treasurer and the Editor.

On behalf of the Clay Minerals Group Dr. D. M. C. MacEwan inquired whether the Council would be prepared to support an appeal to industry for a grant in support of the proposed monograph on clay mineral data. It was agreed that in the first place an application should be made to

the Royal Society for a grant from the Parliamentary Grant-in-aid for Scientific Publications.

A report was received from the Committee on the Composition and method of election of the Council, and the proposed new Bye-laws relating to this were further discussed. After approval of the proposed new Bye-laws, it was agreed to submit them to the Society at a Special General Meeting to be held on June 24.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., April 1, 1948, *at* 5 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

Thirty-two Members and twelve Visitors were present.

The elections of twelve Ordinary Members of the Society provisionally made by the Council were confirmed.

The following exhibits were shown:

1. Changes in the optical orientation of ganophyllite on dehydration: by Dr. W. Campbell Smith (vol. 28, p. 348).

2. Structure model of α-quartz: by F. B. Johnson on behalf of Signals Research and Development Establishment (Ministry of Supply), Christchurch, Hants, demonstrated by Miss D. B. Fletcher.

3. Anorthoclase crystals from Mount Kenya: by Prof. F. E. Zeuner.

The following papers were read and discussed:

1. The identification of minerals in boiler deposits. Examples of hydrothermal synthesis in boilers: by Dr. L. M. Clark (communicated by Dr. F. A. Bannister) (vol. 28, p. 359).

2. Davidite from Tete, Portuguese East Africa: by Dr. F. A. Bannister.

3. A simple method of determining the refractive indices of the felspars: by Miss O. Robins (communicated and read by Dr. A. K. Wells) (vol. 28, p. 367).

4. Catalogue of topographical mineralogies and regional bibliographies: by Dr. L. J. Spencer (vol. 28, p. 303).

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., June 24, 1948, *at* 3.15 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

The following were provisionally elected Ordinary Members of the

Society: Stanley Hay Umphray Bowie, Ralph Jerome Holmes, Andrew Leigh Johnson, Walter David Keller, Stoyan Pavlović, Benjamin Martin Shaub, and Helmut Gustav Franz Winkler.

The deaths of Professor A. Lacroix, Honorary Member since 1900 and previously an Ordinary Member from 1885, and H. P. Whitlock were reported.

Reports were received from the Treasurer and the Editor.

The list of Officers and Council for 1948–49 was considered. The following were due to retire: President, Dr. W. Campbell Smith; Ordinary Members of Council, Dr. M. H. Hey, Dr. J. H. Taylor, Dr. A. E. J. Vickers, and Prof. L. R. Wager. The President proposed as his successor Prof. C. E. Tilley and this was unanimously approved. The following were proposed as Ordinary Members of Council: Dr. S. O. Agrell, Mr. H. G. Dines, Mr. G. S. Gowing, and Dr. S. I. Tomkeieff.

The Secretary stated that he had in hand five papers not yet read and that several others had been promised. It was desirable to hold an additional meeting for the reading of some of these papers, and it was agreed to hold such a meeting on October 7 and to postpone the Anniversary Meeting to November 25.

The following dates of meeting in 1949 were provisionally agreed: Thursdays, January 27, March 31, June 23, and November 3.

SPECIAL GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., June 24, 1948, *at* 5 *p.m.*

The following alterations of the Bye-laws relating to the composition and method of election of the Council were unanimously approved:

11. The Society shall be governed by a Council consisting of a President, two Vice-Presidents, a General Secretary, a Foreign Secretary, a Treasurer, an Editor of the Journal, and twelve Ordinary Members, who shall be elected annually, together with the preceding President,* who shall be entitled to remain on the Council for three years from the date of his retirement from the Chair, and ex officio the Managing Trustees of the Society.

*Presidents who were elected previous to the date of the passing of this Bye-law may remain Life Members of the Council as provided in the original Bye-laws revised in 1931 and 1940.

14. The Officers, Vice-Presidents, Foreign Secretary, and Ordinary Members of Council shall be elected annually at the Anniversary Meeting, which shall be held in November each year or as near thereto as may be convenient.

15. Four Ordinary Members of Council shall retire annually at the Anniversary Meeting, and shall not be eligible for re-election as such until the next Anniversary Meeting.

16. At or before the end of June in each year the Council shall prepare a list of Members nominated by the Council as Officers, Vice-Presidents, Foreign Secretary, and as Ordinary Members of Council for the next ensuing year.

17. The list of Members nominated by the Council in accordance with the preceding Bye-law shall be sent by the General Secretary to every Member of the Society at his last known address not later than the next ensuing July 20.

18. Additional nominations may be made by Members of the Society, not being Members of the Council, subject to the following conditions:

(*a*) All such nominations must be received by the General Secretary not later than the next ensuing September 1, and no nomination received after that date shall be valid.

(*b*) Every Candidate, whether proposed as an Officer, Vice-President, Foreign Secretary, or Ordinary Member of Council, must be proposed by not less than eight Members of the Society, not being Members of the Council, who must all sign the Candidate's Nomination Form, which must be accompanied by a statement signed by the Candidate that he accepts nomination.

(*c*) No Member of the Society may propose more than four Candidates.

(*d*) No Candidate may be nominated for more than one position on the Council.

(*e*) No Member of the Society whose name has been entered on the list of Candidates nominated by the Council for Ordinary Membership of Council may accept nomination under this Bye-law for any other position on the Council.

19. If through death or for any other reason a vacancy should be created on the list circulated by the Council, the Council shall have power to fill the vacancy thus created, and shall notify the Members of the Society of this change as soon as possible, in any case not later than on the notice convening the Anniversary Meeting.

20. If no more than nineteen Candidates are nominated and the nominations be in accordance with the provisions of these Bye-laws, the Candidates shall be declared duly elected at the next ensuing Anniversary Meeting.

21. If more Candidates are nominated for any of the positions on the Council set out in Bye-law 11 than there are places to be filled, then an election by Ballot shall be held.

22. In the event of an election by Ballot being required, the General Secretary shall issue to every Member of the Society at his last known address not later than the next ensuing September 30 a voting-paper containing the names of all Candidates duly nominated in accordance with the provisions of these Bye-laws, which list shall show the Office or other position on the Council for which each Candidate has been nominated.

23. No vote may be cast for any Candidate whose name is not entered on the voting list and no member may vote for more than nineteen Candidates in all.

24. Every Member voting shall place his voting-paper when duly completed in an envelope, which shall be closed and placed in another envelope for return to the scrutineers of the Ballot. The outer envelope must also be closed, must be inscribed 'Voting-Paper', and must bear the name of the Member voting, but no indication of the Voter's identity may appear on the voting-paper or on the inner envelope. Members voting may return their voting-papers enclosed in the manner prescribed to the General Secretary beforehand by post or other means or may deliver them to him at the commencement of the Anniversary Meeting.

25. As soon as may be convenient after the opening of the Anniversary Meeting the Chairman of the Meeting shall appoint from amongst the Members present two Members, not being Members of the Council or Candidates for election to the Council, to act as Scrutineers of the Ballot, to whom the outer envelopes containing the voting-papers received by the General Secretary shall be forthwith delivered unopened and when this has been done the Ballot shall be declared closed.

26. Upon completion of their examination of the voting-papers the Scrutineers shall present to the Chairman of the Meeting a list showing the result of the Ballot, and if the list be in accordance with the provisions of these Bye-laws, the Chairman shall declare the new Council to have been duly elected and to be constituted in accordance with the provisions of these Bye-laws.

27. If, however, any difficulty should arise in constituting the new Council in conformity with the provision of these Bye-laws from the list returned by the Scrutineers owing to an equality of votes or otherwise, the decision of the Chairman, who shall have a casting vote, shall be final, provided always that in no case shall the Chairman declare elected more than fifteen Members who were Members of the retiring Council.

32. Any Member of the Society appointed a Managing Trustee shall be appointed in the first instance for a period of 5 years, and he shall be eligible thereafter for reappointment for such further periods of 5 years at any one time as the Council may determine.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., June 24, 1948, *at* 5.15 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

Thirty-nine Members and seven Visitors were present.

The elections of seven Ordinary Members of the Society provisionally made by the Council were confirmed.

The following exhibits were shown:

1. Illustrated descriptions of two early microscopes: by Dr. A. F. Hallimond.

2. Harkerite from Skye, a new boron mineral: by Prof. C. E. Tilley.

The following papers were read and discussed:

1. The application of phase-contrast microscopy to mineralogy and petrology: by Dr. F. Smithson (vol. 28, p. 384).

2. The lattice parameters and interplanar spacings of some artificially prepared melilites: by Dr. K. W. Andrews (communicated by Dr. F. A. Bannister) (vol. 28, p. 374).

3. The densest and least dense packing of equal spheres: by Mr. S. Melmore (vol. 28, p. 479).

4. On the delimitation of diorite and gabbro and related rocks: by Mr. S. E. Ellis (communicated by Dr. W. Campbell Smith) (vol. 28, p. 447).

5. Pyroxene from the Squilver dolerite, south Shropshire: by Mr. F. G. H. Blyth (vol. 28, p. 380).

6. Ships' loadstones: by Mr. C. E. N. Bromehead (vol. 28, p. 429).

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., October 7, 1948, *at* 4 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

The following were provisionally elected Ordinary Members of the Society: George Baker, George Bisson, Gabriel Martin Cardoso, Carl Wilhelm Erich Correns, Joaquin Folch-Girona, Jack Joseph Frankel, Gilbert Alec Garceau, André George Gregory, John Ewen Troup Horne, Eric Robert Levett, Ronald Walter Brown Nurse, Ernest Hugo Charles Rutland, and André Philippe Sandrea.

The death of M. I. Khan was reported. The resignation of F. E. Studt was accepted.

The Treasurer reported that £250 had been received from the Royal Society (Parliamentary Grant-in-aid for Scientific Publications) towards the cost of publication of a monograph on clay mineral data.

The Secretary drew attention to a decrease in the stock of recent numbers of the Magazine, and it was decided to print a further 50 copies (total 1050) of the December 1948 issue (no. 203) and to review the position for future numbers.

The President commented upon the success of the Society's visit, with foreign guests from the International Geological Congress, to Sir Arthur Russell's collection of minerals at Swallowfield Park, Reading. It was decided that the cost of transport from London be borne by the Society, and that a letter of thanks be sent to Sir Arthur Russell for his generous hospitality.

GENERAL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., October 7, 1948, *at* 5 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

Thirty Members and four Visitors were present.

The elections of thirteen Ordinary Members of the Society provisionally made by the Council were confirmed.

The following papers were read and discussed:

1. On the occurrence of anatase in sedimentary kaolin: by Dr. G. Nagelschmidt, Mr. H. F. Donnelly, and Mr. A. J. Morcom (vol. 28, p. 492).

2. A list of catalogues of meteorite collections: by Dr. L. J. Spencer (vol. 28, p. 471).

3. Clinozoisite from Camaderry Mountain, Co. Wicklow: by Mr. R. W. Johnston (communicated by Prof. C. E. Tilley) (vol. 28, p. 505).

4. An optical and X-ray examination of the basic slag mineral, silicocarnotite: by Dr. D. P. Riley and Mr. E. R. Segnit (vol. 28, p. 496).

5. Two olivines from South African melilite-basalts: by Dr. Morna Mathias (vol. 28, p. 486).

EXCERPT MINUTES OF COUNCIL MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., November 25, 1948, *at* 3.15 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

The following were provisionally elected Ordinary Members of the Society: Yehia Anwar, Frederick Charles Michell Bawden, William Robert Dearman, Mrs. Audrey Mary Brasnett Douglas, Robert Brian Elliott, Peter Godwin Embrey, Paolo Gallitelli, Jack Hartley, George Mueller, Martyn Edward Pease, Eric Harold Timothy Whitten.

The death of E. W. Tunbridge was reported. The resignation of W. F. Fleet was accepted.

Mr. T. Deans and Dr. David Williams were appointed auditors for 1949.

Reports were received from the Treasurer and the Editor. The Secretary reported that the number of copies of the Magazine held in stock for nos. 188–195 (vol. 27, 1945–46) was very low; and it was decided to proceed with the reprinting of no. 194, of which only three copies were left.

The draft Annual Report, that had already been circulated to members of Council, was amended and approved.

GENERAL (ANNIVERSARY) MEETING

Held in the Apartments of the Geological Society, Burlington House, London, W., November 25, 1948, *at* 5 *p.m.*

Dr. W. Campbell Smith, President, in the Chair.

Forty-three Members and nine Visitors were present.

The elections of eleven Ordinary Members of the Society provisionally made by the Council were confirmed.

The General Secretary read the following:

ANNUAL REPORT OF THE COUNCIL FOR 1947–48.

Membership. The total membership on October 31, 1948, was 411 against 351 on October 31, 1947. Sixty-seven new members were elected during the year, but the election of one of these was subsequently declared void. The Society has lost by death its senior Honorary Member, Professor A. Lacroix, and four Ordinary Members. On October 31 the membership consisted of 6 Honorary Members and 405 Ordinary Members of whom 68 are Life Members. The increase in membership during the past twelve months is the greatest in the history of the Society.

Meetings.—The total attendance at the five meetings held during the session 1947/48 was 207 members and 45 guests, giving an average of 50 per meeting. At these meetings 24 papers have been read and 10 exhibits shown.

In connexion with the International Geological Congress and at the very kind invitation of Sir Arthur Russell the Society was able to invite its overseas members to Swallowfield Park, Reading, and give them the opportunity of seeing Sir Arthur's collection.

Clay Minerals Group.—Two meetings have been held in London during the year. The first of these, in January, occupied two sessions at which nine papers were read; the second, during the International Geological Congress, occupied three sessions and about fifteen papers were read. All the meetings were well attended. A second number of the Clay Minerals Bulletin has been issued during the year.

Bye-laws. —The Bye-laws concerning composition fees and the method of election of the Council have been revised.

Publications. Four numbers (199 202) of the *Mineralogical Magazine* have been issued during the year, containing the Proceedings in 14 pages, 18 papers in 217 pages with 11 plates and 45 text-figures, and 725 abstracts in 204 pages.

In consequence of the increase in membership of the Society and subscribers to the Magazine, the present printing of 1000 copies per issue will be increased to 1050 with the December number.

Financial.—The accounts for the calendar year 1947 published in the March number of the Magazine show an excess of assets (exclusive of the stock of the Magazine and the Society's other effects) over liabilities of £3896. 6*s*. 1*d*. against £3417. 2*s*. 6*d*. at the close of 1946. The value of the securities standing in the name of the Custodian Trustee was on December 31, 1947: General Reserve £2839. 1*s*. 6*d*., Miers Memorial Fund £435. 0*s*. 0*d*.

The balances standing to the credit of the General Purposes Account at the close of 1947 amounted to £737. 9*s*. 0*d*. against £494. 18*s*. 2*d*. at the close of 1946, and that standing to the credit of the Miers Memorial Fund £30. 15*s*. 2*d*. against £15. 4*s*. 2*d*. The outstanding liabilities at the close of 1947 amounted to £283. 1*s*. 3*d*. against none at the close of 1946.

The receipts from Entrance Fees and Annual Subscriptions have been satisfactory. For the period January 1 to October 31 the marked increase which was shown in 1947 has been maintained for the corresponding period in 1948. The effect of raising substantially the charges for the Magazine which came into force on June 1, 1947, is seen in the receipts from the Society's publisher for sales of the *Mineralogical Magazine* during the year April 1, 1947, to March 31, 1948, which

were £902. 15*s*. 0*d*. against £361. 5*s*. 10*d*. for the previous year. The net receipts from sales by the Custodian of the stock of the Magazine in 1947 totalled £109. 2*s*. 3*d*. against £39. 10*s*. 0*d*. in 1946. The gross receipts from this source from January 1 to October 31, 1948, were £3. 1*s*. 0*d*.

Although the present financial position of the Society is satisfactory, it will require careful watching in the immediate future. In 1947 the outgoings on the Magazine totalled £710. 13*s*. 3*d*. against £699. 18*s*. 1*d*. in 1946. The expenditure on the four numbers (nos. 196–199) due for issue in 1947 was £983. 17*s*. 6*d*. against £695. 18*s*. 1*d*. in respect of the previous four numbers. Three numbers have been issued since the beginning of the current year which have cost £693. 12*s*. 1*d*. inclusive of the cost of authors' separate copies, and it is estimated that the figure for the four numbers issued and to be issued in 1948 will be about £940.

The Society has received through the Royal Society a grant of £250 from the Parliamentary Grant-in-aid for Scientific Publications for the purpose of producing a monograph of X-ray and other data of clay minerals.

Composition Fees totalling £231. 0*s*. 0*d*. were received during 1947. Since January 1 further Composition Fees totalling £99. 5*s*. 0*d*. have been received. All have been invested except £9. 13*s*. 0*d*.

The balances at Bankers, in the Post Office Savings Bank, and in hand on October 31 were: General Purposes Account £1150. 18*s*. 6*d*., Miers Memorial Fund £34. 16*s*. 2*d*., Publication Grant from Royal Society £250. 0*s*. 0*d*. The bills for the March, June, and September numbers of the Magazine together with those for authors' separate copies have been paid. The outgoings for the remainder of the year are estimated at approximately £340 and the receipts at approximately £95. On this basis the cash available for future use (exclusive of that standing to the credit of the Miers Memorial Fund and the Royal Society Grant) at the close of the year should be about £905 against £737. 9*s*. 0*d*. brought in from 1947.

On the proposal of Dr. K. C. Dunham, seconded by Mr. P. M. Game, the adoption of the Annual Report of the Council was carried unanimously.

A vote of thanks to the Geological Society for the use of its apartments was proposed by Mr. P. A. Sabine and seconded by Mr. B. W. Anderson, and carried with acclamation.

The President read the following list of Officers and Council elected for the session 1948–49, and invited the new President to take the Chair. In doing so he thanked the Officers, Council, and Members for their support during his term of office which had been a memorable and happy one to him.

President. Prof. C. E. Tilley, B.Sc., Ph.D., F.R.S.

Vice-Presidents. Mr. C. E. N. Bromehead, B.A., F.G.S.; Mr. B. W. Anderson, B.Sc., F.C.S.

Treasurer. Dr. A. J. Bull, M.Sc., Ph.D., F.Inst.P., F.G.S.

General Secretary. Dr. G. F. Claringbull, B.Sc., Ph.D., F.G.S.

Foreign Secretary. Mr. F. N. Ashcroft, M.A., F.G.S., F.C.S.

Editor of the Journal. Dr. L. J. Spencer, C.B.E., M.A., Sc.D., F.R.S.

[*Continued on p. lxxx*

ABSTRACT OF RECEIPTS AND PAYMENT

GENERAL PU

Receipts	£	s.	d.	£	s.
By Balances brought in from 1947—					
1. At Bankers on Current Account	252	12	4		
2. In Post Office Savings Bank	482	17	10		
3. In Office	1	18	10		
				737	9
Entrance Fees				59	17
Annual Subscriptions—					
1. Current year	295	1	0		
2. Arrears	6	6	0		
3. Advance	30	9	0		
				331	16
Sale of Publications—					
1. By Publisher, April 1, 1947–March 31, 1948. Sales at subscription rate £874. 0*s*. 0*d*. and other sales £400. 11*s*. 0*d*. less trade discount and commission £352. 0*s*. 4*d*. and distribution charges £28. 9*s*. 1*d*.	894	1	7		
2. By Custodian of Stock, 1948, £95. 4*s*. 0*d*. less distribution costs £2. 13*s*. 3*d*. and cost of second-hand copies purchased £27. 9*s*. 0*d*.	65	1	9		
				959	3
Advertisement Fees £10. 14*s*. 4*d*. less commission £2. 0*s*. 11*d*.				8	13
Interest on Investments, including Income Tax recovered—					
1. £1685 3½% War Loan, 1952 and after	58	19	6		
2. £105 4% Consolidated Loan, 1957	4	4	0		
3. £100 4% Funding Loan, 1960–90	4	0	0		
4. £146. 10*s*. 0*d*. 3% War Loan, 1955–59	4	7	8		
5. £255 3% Defence Bonds	7	13	0		
6. £250 3% Savings Bonds, 1960–70	7	10	0		
7. £100 3% Savings Bonds, 1965–75	1	10	0		
8. £275 2½% Consolidated Stock	4	7	1		
				92	11
Transfers from Miers Memorial Fund				30	0
Donations—					
1. Royal Society (Parliamentary Publications Grant)	250	0	0		
2. Other Donations	31	12	6		
				281	12
Clay Minerals Group, notices to non-members				11	12
Small items				0	0
Interest on Post Office Savings Bank deposit				18	6
				£2531	1

)CIETY

'R THE YEAR ENDING DECEMBER 31, 1948

SES ACCOUNT

Payments	£ s. d.	£ s. d.
Printing and distributing the *Mineralogical Magazine*, Nos. 199–203 (Oxford University Press)—		
1. Printing	999 0 3	
2. Illustrations	110 2 5	
3. Authors' Separate Copies (Nos. 198–202)	58 19 11	
4. Distribution Charges	24 4 9	
		1192 7 4
Reprinting of *Mineralogical Magazine* No. 192		76 12 6
Miscellaneous Printing, not annual		36 17 0
Printing and distributing Notices of Meetings and other annual printing		38 18 7
Expenses at Meetings—		
1. Operator of Lantern	3 3 0	
2. Porter	4 15 0	
3. Tea	3 9 5	
		11 7 5
Editor's Honorarium		50 0 0
Care of Stock of Publications, J. Mugford		1 1 0
Clay Minerals Group, Expenses		13 5 3
International Geological Congress—		
1. Fees of Delegates	4 0 0	
2. Excursion to Swallowfield Park	25 4 0	
3. Secretary's expenses	1 0 0	
		30 4 0
Bank Charges, Exchange Losses, and other Small Items		1 19 8
Postage—		
1. Hon. Secretary	8 12 6	
2. Hon. Treasurer	6 0 0	
		14 12 6
Balances, December 31, 1948—		
1. At Bankers on Current Account	53 15 3	
2. In Post Office Savings Bank	1001 3 10	
3. In Office	8 16 11	
		1063 16 0
		£2531 1 3

mined and compared with Vouchers and found correct.

T. DEANS
D. WILLIAMS } Auditors.

A. J. BULL, Hon. Treasurer.

uary 21, 1949.

MIERS MEMORIAL FUND, YEA

Receipts	£	s.
By Balance brought in from 1947—		
At Bankers on Current Account	30	15
Donation	21	0
Interest on Investments, including Income Tax recovered	13	1
	£64	16

COMPOSITION FEES (INVESTMENT) ACCOUN

Receipts	£	s.
By Balance brought in from 1947—		
At Bankers on Current Account	10	14
Composition Fees received during year	169	5
	179	19

Note:—The total sum received as Composition Fees, of which £79. 13*s.* 0*d.* was then uninvested, was on December 31, 1948, £1796. 15*s.* 0*d.*

BALANC

DECEM

Liabilities	£	s.	d.	£	s
To General Funds:—					
Annual Subscriptions received in advance—					
Received in 1948 for 1949	27	6	0		
Received in 1948 for 1950	1	1	0		
Received in 1948 for 1951	1	1	0		
Received in 1948 for 1952	1	1	0		
				30	9
Sundry Creditors—					
Oxford University Press (Printers)—					
1. For Authors' separate copies from No. 203 of the *Mineralogical Magazine* estimated at	12	0	0		
2. For Distribution Charges for No. 203 of the *Mineralogical Magazine* estimated at	6	0	0		
				18	0
Special Funds:—					
Miers Memorial Fund				469	16
Excess of Assets over Liabilities				4697	
				£5215	

DING DECEMBER 31, 1948

PAYMENTS	£	s.	d.
Balance, December 31, 1948—			
Provision of plates in Nos. 201 and 202	30	0	0
At Bankers on Current Account	34	16	2
	£64	16	2

amined and compared with Vouchers and found correct.

T. DEANS
D. WILLIAMS } Auditors.

uary 21, 1949. A. J. BULL, Hon. Treasurer.

EAR ENDING DECEMBER 31, 1948

PAYMENTS	£	s.	d.
Investments; purchase of £100 3% Savings Bonds	100	6	0
Bankers on Current Account	79	13	0
	179	19	0

T. DEANS
D. WILLIAMS } Auditors.

uary 21, 1949. A. J. BULL, Hon. Treasurer.

HEET

1948

ASSETS	£	s.	d.	£	s.	d.
General Funds:—						
nvestments at Broker's Valuation—						
1. £1685 3½% War Loan, 1952 and after, at 103 7/16%	1742	18	6			
2. £105 4% Consolidated Loan, 1957, at 109 5/16%	114	15	7			
3. £100 4% Funding Loan, 1960–90, at 114¼%	114	5	0			
4. £146. 10*s.* 0*d.* 3% War Loan, 1955–59, at 105 13/16%	155	0	3			
5. £255 3% Defence Bonds at par	255	0	0			
6. £250 3% Savings Bonds, 1960–70, at 103 5/16%	258	11	10			
7. £100 3% Savings Bonds, 1965–75, at 102 7/8%	102	17	6			
8. £275 2½% Consolidated Stock at 80%	220	0	0			
				2963	8	8
Sundry Debtors—						
Oxford University Press (Publishers)						
(*a*) For sales of publications, April 1–December 31, 1948	609	15	3			
(*b*) For Advertisement Fees, April 1–December 31, 1948	14	4	6			
				623	19	9
Entrance Fees in Arrear				4	4	0
Annual Subscriptions in Arrear, estimated to realize				10	10	0
Balances—						
1. At Bankers on Current Account	133	8	3*			
2. In Post Office Savings Bank	1001	3	10			
2. In Office	8	16	11			
				1143	9	0
Special Funds:—						
Miers Memorial Fund—						
Investments						
£435 3% Defence Bonds at par	435	0	0			
Balance—						
At Bankers on Current Account	34	16	2			
				469	16	2
				£5215	7	7

* Including £79. 13*s.* 0*d.* being uninvested balance standing to the credit of the Composition Fees (Investment) Account

Note:—The value of the periodicals retained after the disposal of the Library, that of the old stock of Publications, and that of the Society's other effects are not included in the ive Assets.

uary 27, 1949. A. J. BULL, Hon. Treasurer.

Ordinary Members of Council.

Mr. T. Deans, M.A., F.G.S.
Dr. W. A. Deer, M.Sc., Ph.D., F.G.S.
Mrs. Kathleen Lonsdale, D.Sc., F.R.S.
Mr. R. C. Spiller, M.A.
Dr. G. W. Brindley, M.Sc., Ph.D., F.Inst.P.
Dr. F. Dixey, O.B.E., D.Sc., F.G.S.
Prof. L. Hawkes, D.Sc., F.G.S.
Prof. W. Q. Kennedy, D.Sc., Ph.D., F.G.S.
Dr. S. O. Agrell, B.A., Ph.D., F.G.S.
Mr. H. G. Dines, A.R.S.M., F.G.S., A.I.M.M.
Mr. G. S. Gowing, M.A., F.G.S.
Dr. S. I. Tomkeieff, M.Sc., D.Sc., F.G.S.

Professor Tilley, on taking the Chair, thanked the Members for the honour done to him. A vote of thanks to the retiring President and Ordinary Members of Council was proposed by Sir Arthur Russell and seconded by Mr. A. F. Seager.

The following exhibits were shown:

1. Examples of the contact-film method of opaque mineral identification: by Mr. F. M. Nakhla and Dr. David Williams.

2. A faceted specimen of the new artificial (Linde) rutile showing a brilliant display of prismatic colours: by Mr. B. W. Anderson.

The following papers were read and discussed:

1. Baryte-bearing nodules from the Middle Lias of the east Midlands: by Dr. J. H. Taylor.

2. X-ray studies of halloysite and metahalloysite. Part I. The structure of metahalloysite, an example of a random layer lattice: by Dr. G. W. Brindley and Dr. K. Robinson (vol. 28, p. 393).

3. X-ray studies of halloysite and metahalloysite. Part II. The transition of halloysite to metahalloysite in relation to relative humidity: by Dr. G. W. Brindley and Dr. J. Goodyear (vol. 28, p. 407).

4. X-ray studies of halloysite and metahalloysite. Part III. Effect of temperature and pressure on the transition from halloysite to metahalloysite: by Dr. G. W. Brindley, Dr. K. Robinson, and Dr. J. Goodyear (vol. 28, p. 423).

The following paper was taken as read:

Crystallization of basaltic magma as recorded by variation of crystal-size in dikes: by Dr. H. G. F. Winkler.